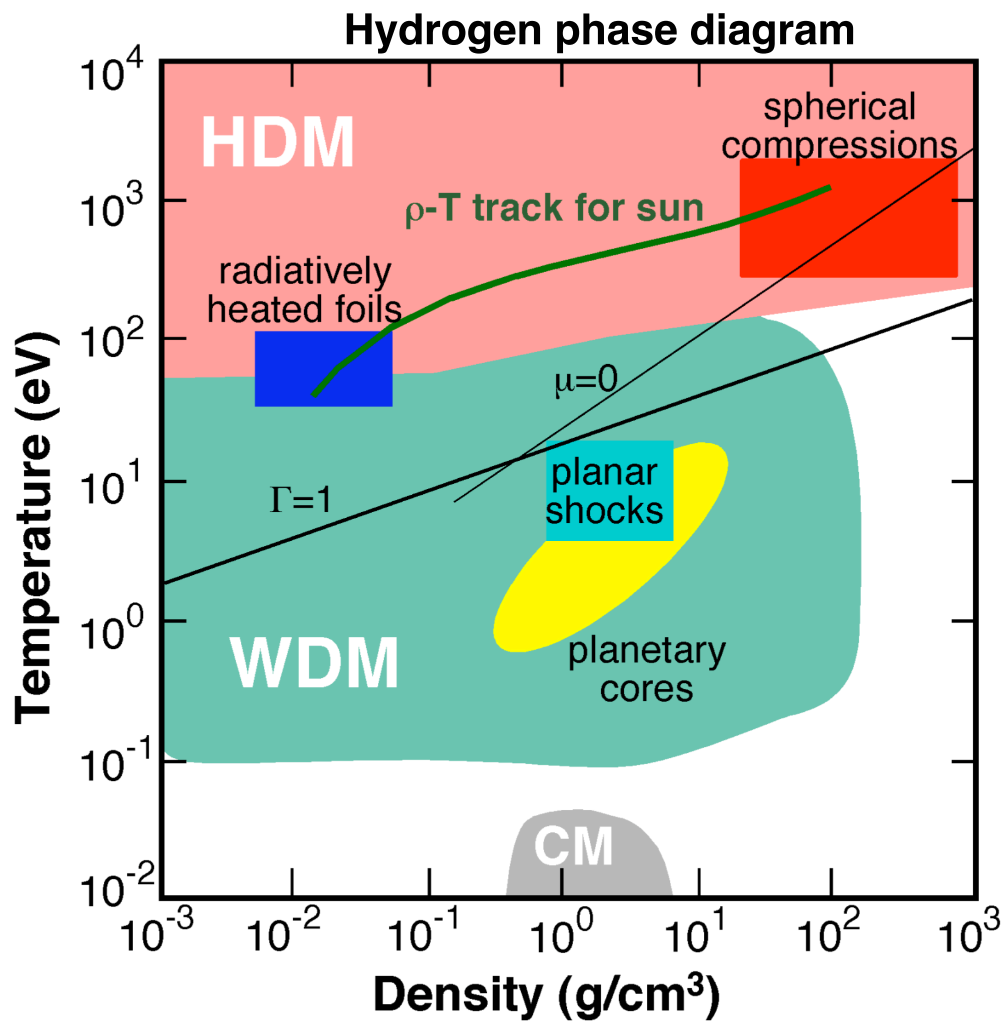
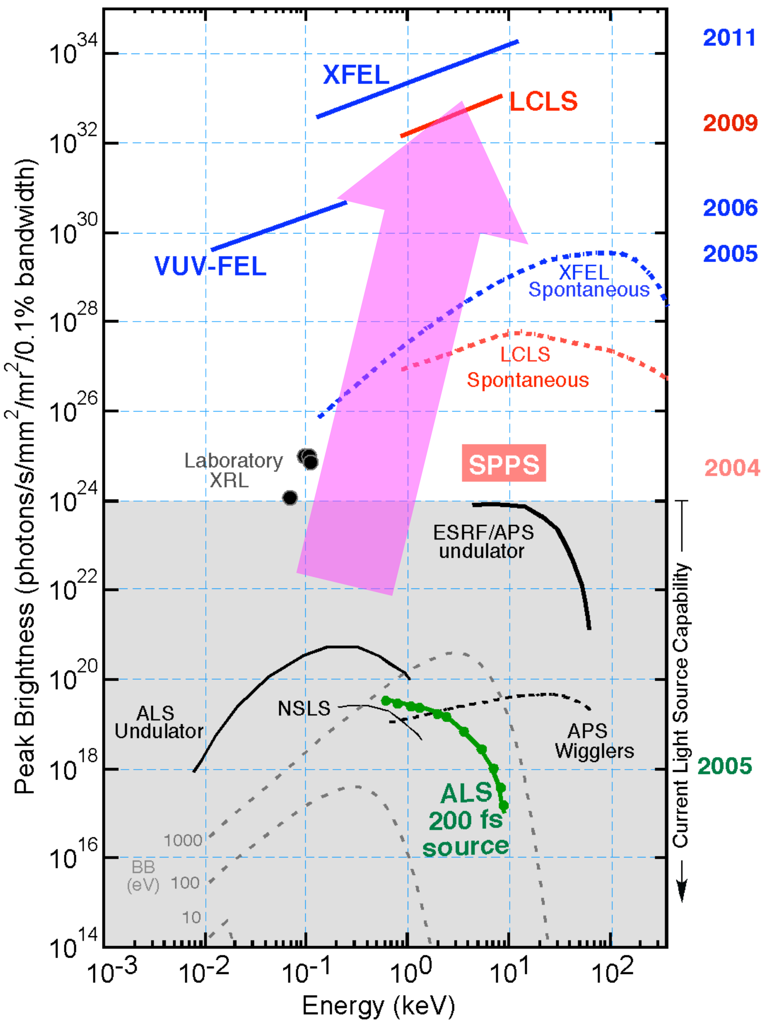
HEDS covers a vast region in T-ρ phase space and numerous physical regimes

- **Hot Dense Matter (HDM):**
  - Supernova, stellar interiors, accretion disks
  - Plasma devices: laser produced plasmas, Z-pinches
  - Directly and indirectly driven inertial fusion experiments
- **Warm Dense Matter (WDM):**
  - Cores of large planets
  - Systems that start solid and end as a plasma
  - X-ray driven inertial fusion experiments



Xray free-electron lasers, XFELs, are well suited to HEDS research

- Intense (10^{12} photons), short pulse (~50 fs), tunable sources



Ultrashort pulses are useful …

- to create HED states of matter
- to probe the highly transient behavior of HED states

High photon energy is required…

- to heat the target volumetrically and, thus, minimize gradients
- to directly probe the high densities

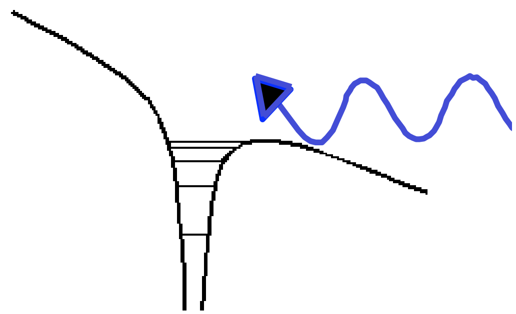
High photon number is useful…

- to make single-shot data feasible

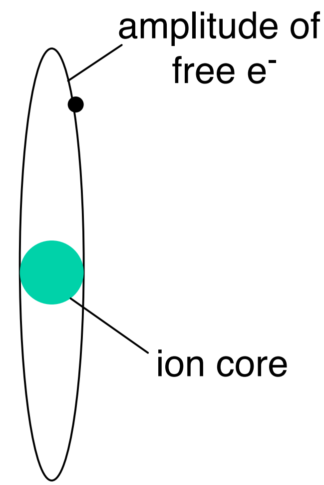
Intense sub-ps x-ray sources are essential to make progress in HED regime

High intensity XFELs interact with cold atoms predominantly by photoionization

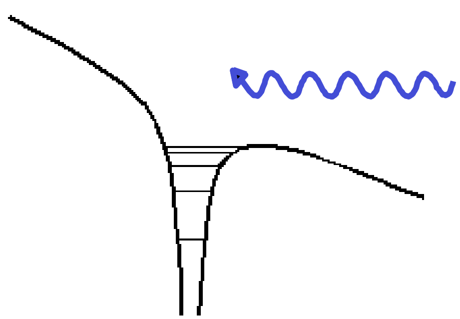
- **Visible/IR laser-atom process at $I_L \geq 10^{14}$ W/cm^2**



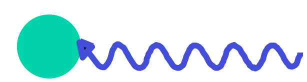
- Field modulates the atomic potential at visible laser frequency
- Outer e^- has time to tunnel free:
  - $2U_p > I_p$ where $U_p \sim (I_L\lambda^2)$
- Strong interaction between free e^- and ion core is of interest



- **FEL-atom process at $I \geq 10^{14}$ W/cm^2**



- Field modulates the atomic potential at x-ray laser frequency
- e^- does not have time to tunnel free
- Important processes are with deeply bound core e^-



FEL-atom interactions is straightforward as the interaction parameter $I\,\lambda^2$ is small

XFELs propagate through dense ionized matter without reflection

- Classical dispersion relation propagation for light through ionized matter :

$$\frac{k_L^2 c^2}{\omega_L^2} = 1 - \frac{\omega_p^2}{\omega_L^2}$$

k_L, ω_L are laser wavenumber and frequency

$\omega_p = \sqrt{4\pi n_e e^2 / m_e}$ = plasma frequency

  - When $\omega_L = \omega_p$ the light will be reflected at the critical density, n_{cr},

$$n_{cr} = \frac{m_e \omega_L^2}{4\pi e^2} \cong \frac{10^{27}}{\lambda_L^2 (nm)}$$

  - For the visible/IR lasers, $\lambda_L \geq 250$ nm, $n_{cr} \leq 1.6\text{x}10^{22}\ e^-/cm^3$
    - For solid density matter, even fractionally ionized, the laser is reflected
  - For the XFELs, $\lambda_L \leq 20$ nm, $n_{cr} \geq 2.5\text{x}10^{24}\ e^-/cm^3$
    - For solid density matter the laser propagates.

XFELs provide for probing and heating solid density matter

XFELs provide a rapid sub-ps source to heat or probe matter. Determine the state of the matter

- 1st requirement is a predictive capability for charge state distribution ⇒
  - Use **FLYCHK** developed by H.-K. Chung

- Simple, fool-proof tool needed to help experimentalist design diagnostics

- General tool applied to any atom under any condition

- Compact module for inclusion in macroscopic codes:
  Hydrodynamics, PIC (Particle-in-cell) and radiation transport…

- Initial accurate estimate of ionization distributions necessary for building more sophisticated kinetics model

H.-K. Chung

Populations can be derived for LTE and non-LTE conditions

- First, in LTE we apply the equilibrium relations of statistical mechanics & thermodynamics at the local T, n
  - This implies $T_R = T_e = T_i$
  - Use Boltzmann relation to get relative level populations,

$$\frac{n_U}{n_L} = \frac{g_U}{g_L}\exp(-\Delta E_{UL}/T)$$

  - Use Saha-Boltzmann to get relative ion stage populations,

$$\frac{n_i}{n_{i+1}n_e} = \frac{g_i}{g_{i+1}g_e}\exp(-\chi_{i,i+i}/T) \quad \text{where } g_e = \frac{2(2\pi mkT)^{3/2}}{\hbar^3}$$

- Second, when the system is non-LTE, i.e., LTE conditions do not exist, we need to solve for the populations using the rates for "all" the process. The rate equations for all levels are given by:

$$\frac{dn_i}{dt} = n_i\left[\sum_{i''<i}\left(\frac{n_{i''}}{n_i}\right)^*\left(R_{ii''} + C_{i''i}\right) + \sum_{i'>i}\left(R_{ii'} + C_{ii'}\right)\right] - \sum_{i''<i} n_{i''}\left(R_{i''i} + C_{i''i}\right) - \sum_{i'>i} n_{i'}\left(\frac{n_i}{n_{ii'}}\right)^*\left(R_{i'i} + C_{ii'}\right)$$

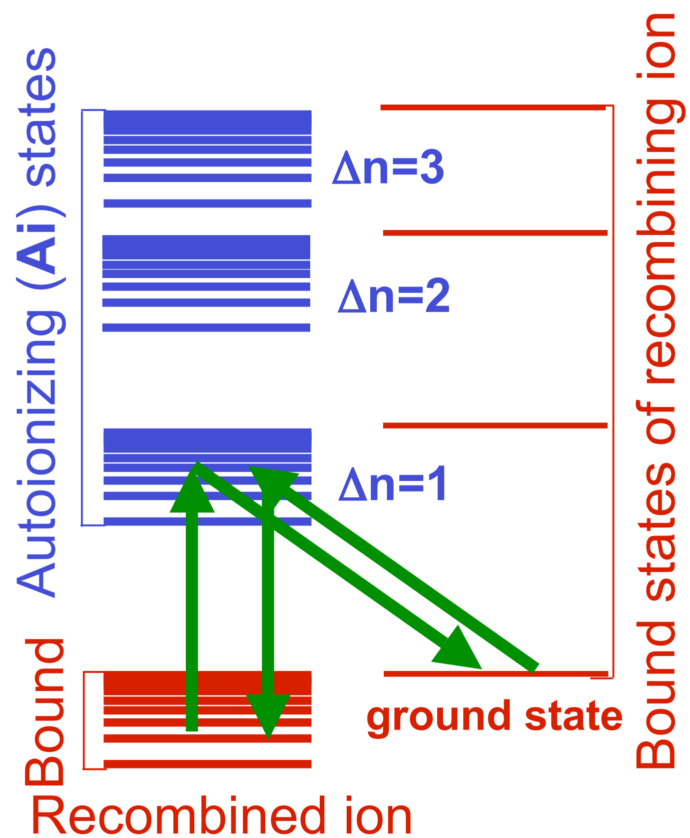
$$R_{LU} = 4\pi\int_{\nu_o}^{\infty}\alpha_{LU}(\nu)J_\nu\frac{d\nu}{h\nu} \qquad R_{UL} = 4\pi\int_{\nu_o}^{\infty}\alpha_{LU}(\nu)\left[\frac{2h\nu^3}{c^2} + J_\nu\right]e^{-h\nu/kT}\frac{d\nu}{h\nu} \qquad \text{where } J_\nu = \oint\frac{d\Omega}{4\pi}I(\vec{r},\vec{n},\nu,t)$$

New method to include EA and DR processes:

1st Essential element for FLYCHK

- Excitation following by Autoionization (EA) and its reverse process Dielectronic Recombination (DR) are critical in many kinetics problems

  - EA/DR processes must be in detailed balance for collision-dominated plasmas
  - EA/DR processes via autoionizing states are modeled within a hydrogenic formalism

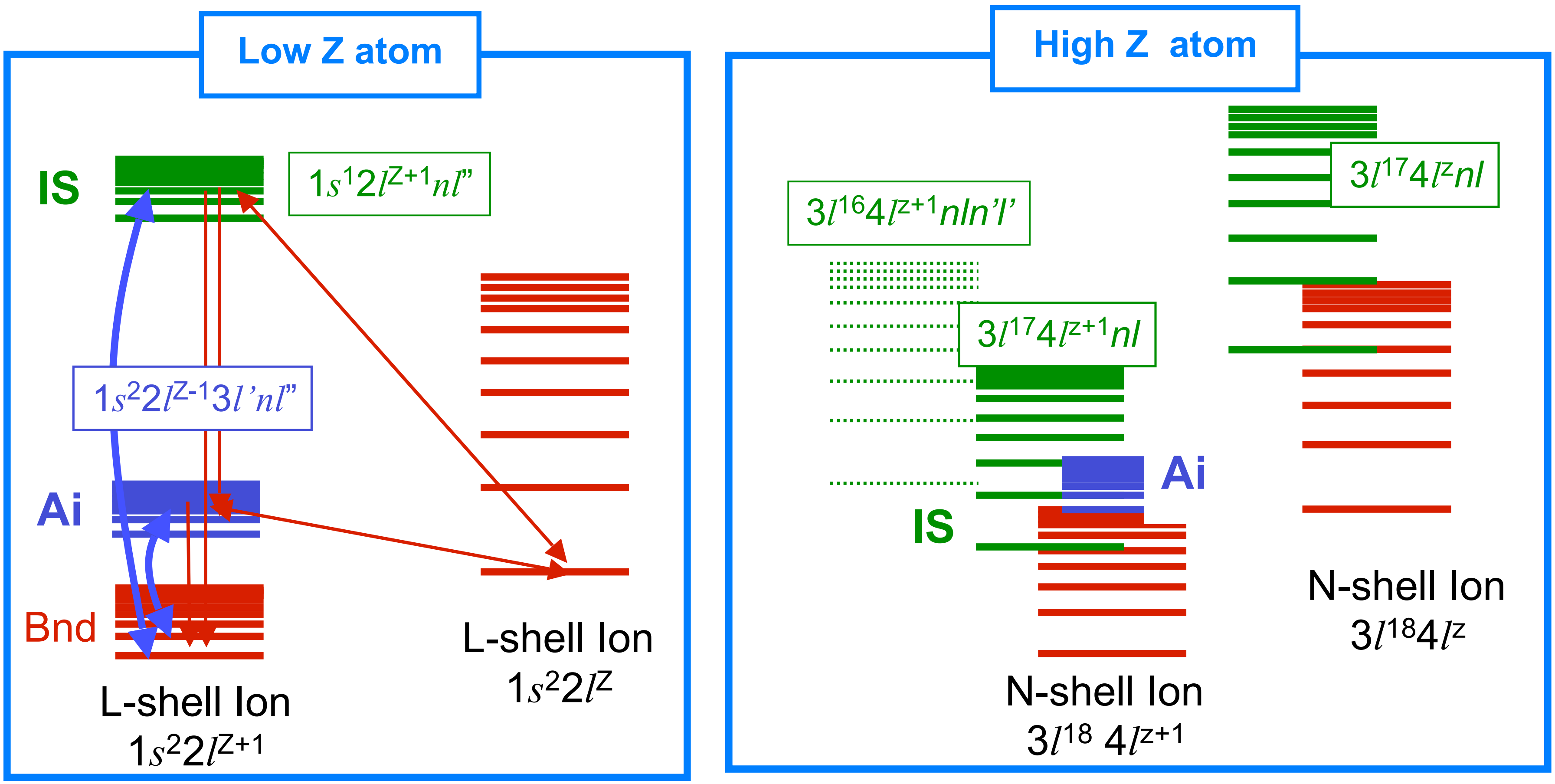


H.-K. Chung

Inner-shell (IS) processes for many-electron ions:

2nd Essential element for FLYCHK

For high-Z atoms, a promotion of inner-shell electrons can lead to states near the continuum limit and hence an accounting of the inner-shell excited states is critical in estimating ionization distributions



H.-K. Chung

2nd requirement is a method to determine temporal distribution of *all* electrons: bound *and* free

- **For highly non-equilibrium fs timescales a new capability is needed**

Ct27: Couples FLYCHK and **Boltzmann Equation solver for Non- LTE electrons**

$$\frac{\partial n_e(\varepsilon)}{\partial t} = \left[\frac{\partial n_e(\varepsilon)}{\partial t}\right]_{Elastic} + \left[\frac{\partial n_e(\varepsilon)}{\partial t}\right]_{\substack{Inelastic \\ \& \, Superelastic}} + \left[\frac{\partial n_e(\varepsilon)}{\partial t}\right]_{Sources} - \left[\frac{\partial n_e(\varepsilon)}{\partial t}\right]_{Sinks} + \left[\frac{\partial n_e(\varepsilon)}{\partial t}\right]_{\substack{Electron- \\ Electron}}$$

where $n_e(\varepsilon) = N_e \varepsilon^{1/2} f(\varepsilon)$ and $\int d\varepsilon f(\varepsilon)\varepsilon^{1/2} = 1$

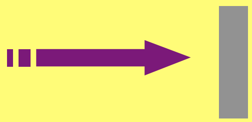
The terms are:

- Elastic losses to phonon (deformation potential) scattering
- Excitation and de-excitation of bound states
- Sources such as photo-ejected and Auger electrons
- Sinks such as three-body, dielectronic, and radiative recombination
- Electron thermalization due to collisions with other electrons

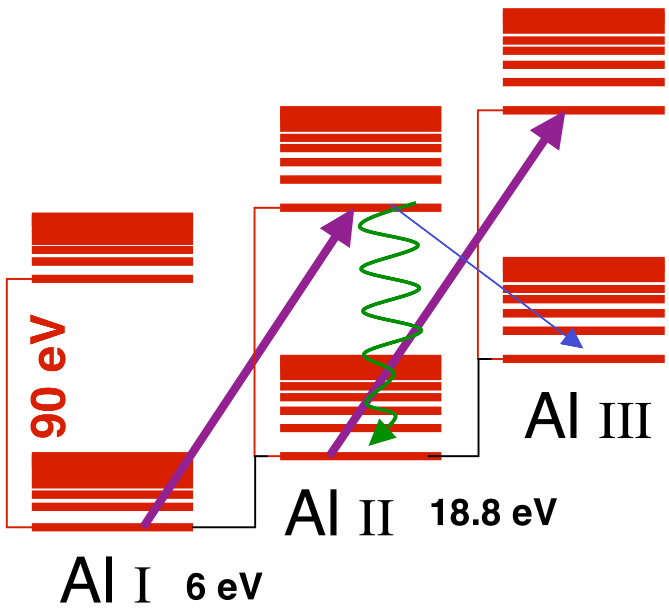
H.-K. Chung

Ct27 has been applied to the FLASH FEL at DESY

- **Problem is to determine the:**
  - Ionization distributions
  - Electron energy distributions
  - Relaxation time scales



- **Experiment:**
  - 200 fs pulse with ΔE/E~0.003
  - 10^{12} 200 eV photons on solid Al 40μ spot
  - Measure incident and transmitted FEL



1. Initially FEL-matter interaction is dominated by inner shell ionization.
2. Auger decay followed immediately
3. Then fractionally ionized solid interacts with e^- through inelastic collisions

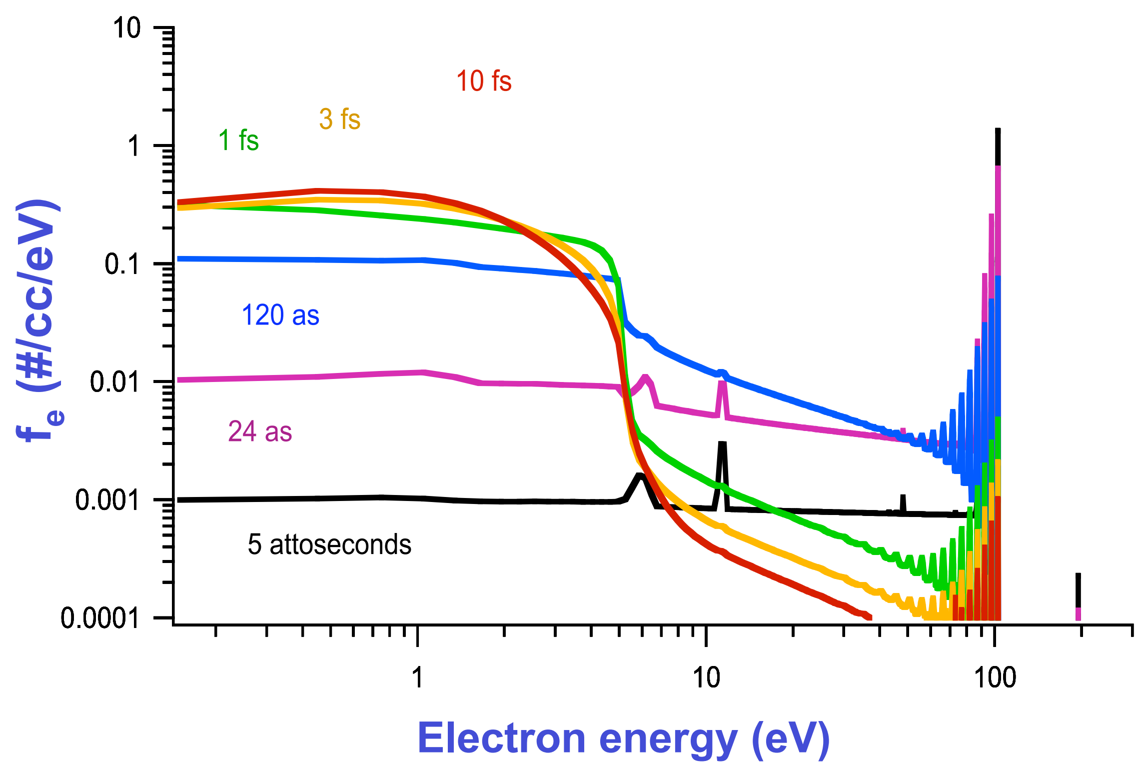
- Timescales for these processes are comparable to the duration of the short pulse x-ray sources, i.e., in the sub-ps regime.

H.-K. Chung

Ct27 indicates that the bulk of the electrons thermalize rapidly

- **Photoelectrons initially produced at 105 eV**
- **e^- thermalize in a few fs due to *inelastic* e^--ion collisions**
- **Average energy of the e^- sharply decreases then rises again**

⇒ XFEL interactions create a broadly thermal solid density



At 5 attoseconds:

$N_e \sim 10^{16} cm^{-3}$: $T_e \sim 65$ eV

$N_i \sim 6 \times 10^{22} cm^{-3}$

e-e elastic ν_{ee} :

Coulomb $\sim 1.4 \times 10^9 \ s^{-1}$

e-i inelastic ν_{ei} :

excitation $\sim 5 \times 10^{16} \ s^{-1}$

ionization $\sim 2 \times 10^{16} \ s^{-1}$

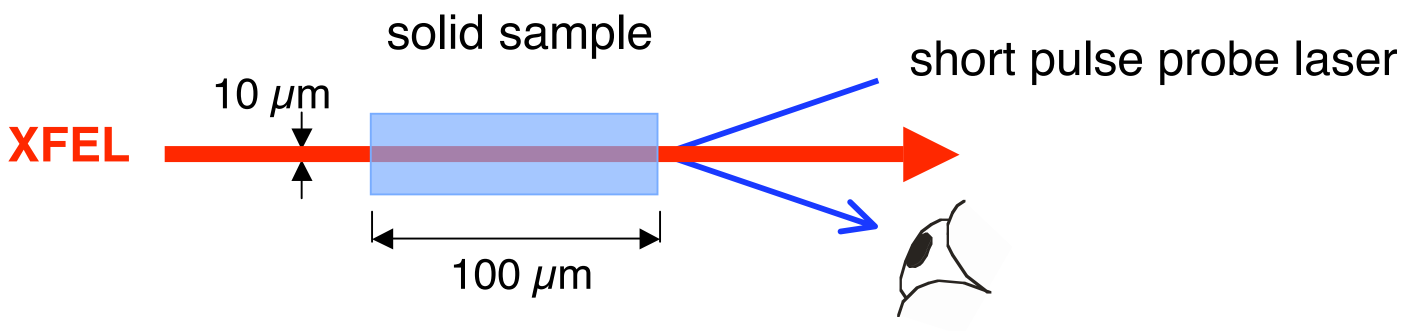
Assumptions

1) No initial solid-state structure

2) No plasma motion

H.-K. Chung

With a sense that the electron distribution will be thermal one *estimate* the sample temperature

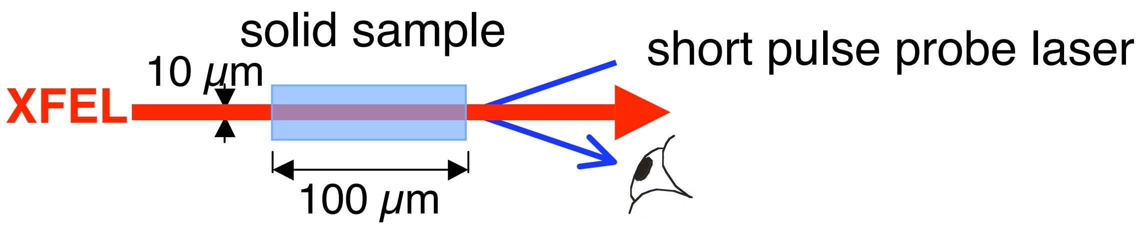


- For a 10x10x100 μm thick sample of Al
  - Ensure sample uniformity by using only 66% of FEL beam energy
  - Equating absorbed energy to total kinetic and ionization energy

$$\frac{E}{V} = \frac{3}{2} n_e T_e + \sum_i n_i I_p^i \quad \text{where } I_p^i = \text{ionization potential of stage } i-1$$

  - Find 10 eV at solid density at 4 Mb with $n_e = 2\times10^{22}$ cm^{-3} and <Z> ~0.3
- Material, rapidly and uniformly heated, releases isentropically

To attain an uniform equilibrium sample two further considerations need to be addressed

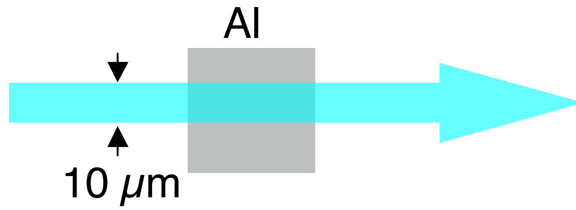


- First, expansion of sample needs to be 'small' on time scale of pulse
  - Surfaces of the sample will move first and an estimate indicates that the expansion on the 1 ps time scale
  - 10 eV surface will move at $v_{expansion} \propto \sqrt{T/m_i} \approx 1.4x10^6 cm/s$
  - In 1 ps the surface therefore moves ~14 nm ⇒ less than 0.1% of sample expands

- Second, **Ct27** treats e^--e^- interactions in detail; but, not ion-e^- elastic interactions
  - i-e^- elastic interaction provide thermalization mechanism for the ions
  - Compared to e^--e^- thermalization time, the i-e^- will be reduced by ~m_e/m_i
    - For the example one estimates T_i to equal T_e in ~3 ps

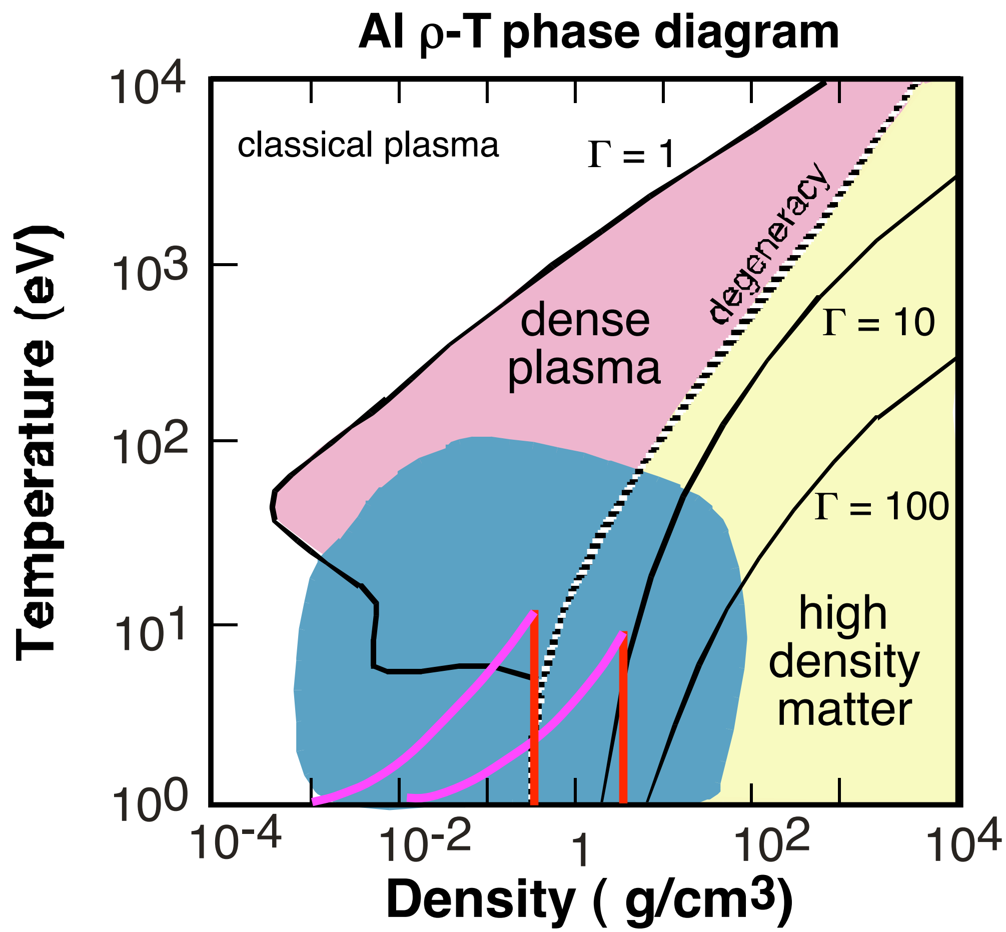
WDM created by isochoric heating will isentropically expand sampling phase space

- Concept is straightforward

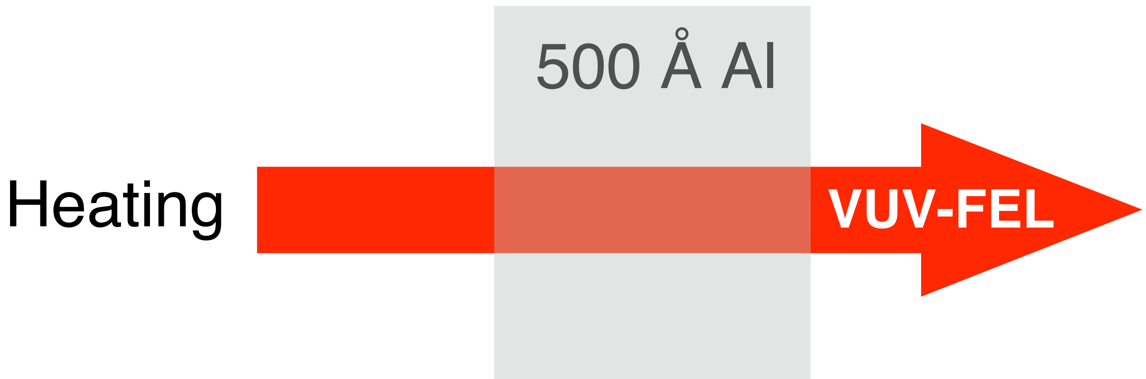


- XFEL can heat matter rapidly and uniformly to create:
  - Isochores (constant ρ)
  - Isentropes (constant entropy)

- Using underdense foams allows more complete sampling
  - Isochores (constant ρ)
  - Isentropes (constant entropy)



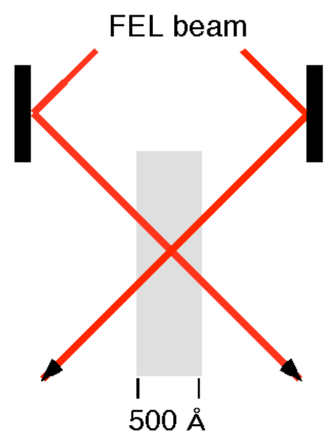
So much for future XFELs: currently we are working to create WDM with FLASH at DESY

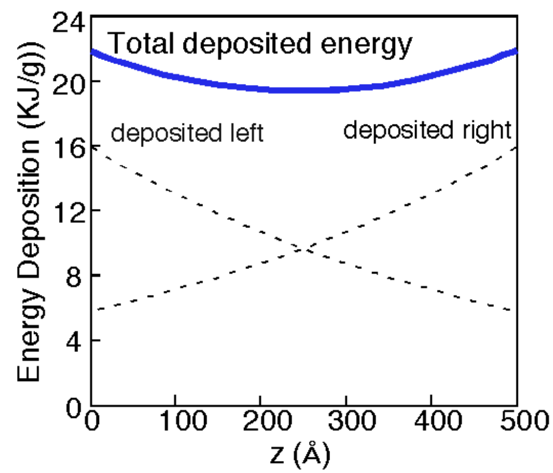


- Isochoric heating
  - 40 fs 60 Å VUV-FEL heats a Al foil 500 Å uniformly
  - $\frac{\mathrm{E}}{\mathrm{V}} = \frac{3}{2} n_e T_e + \sum_i n_i \mathrm{I}_{\mathrm{P}}^{\mathrm{i}} \Rightarrow$ (10^{12} x 200 eV) / Volume = 3/2 (1.7×10^{23}) x 10 eV
    - Volume = Area x 500 Å ⇒ Area = 50 μm spot
    - For 1 eV plasma a 140 μm spot is needed
- Isentropic expansion
  - A optical FDI probe measures the isentropic expansion

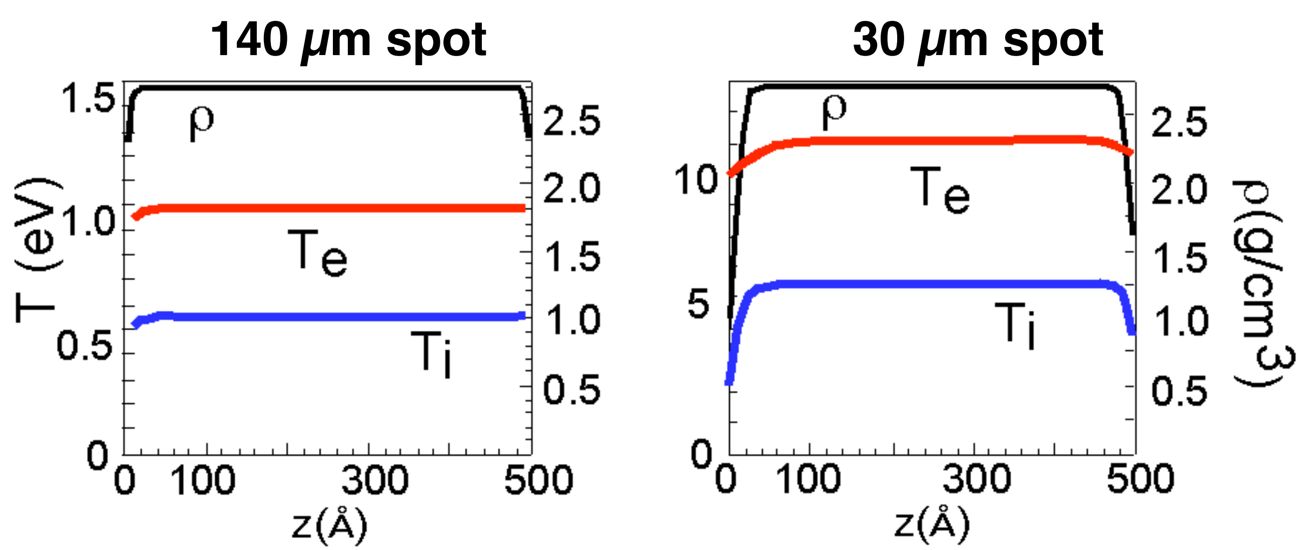
Simulations of FLASH VUV-FEL confirm simple estimates for creating WDM:

- 500 Å Al irradiated by split FLASH with 200 fs pulse width

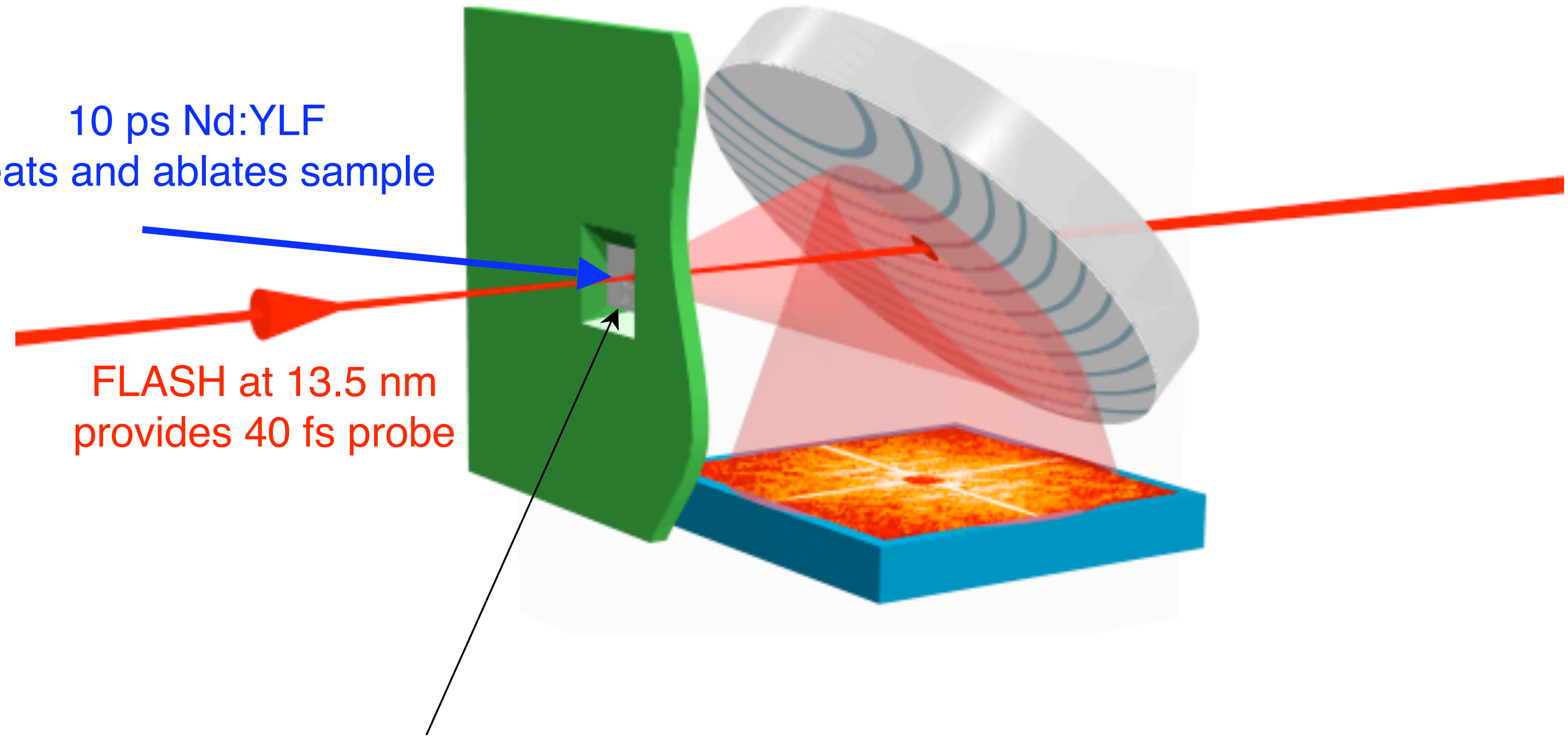




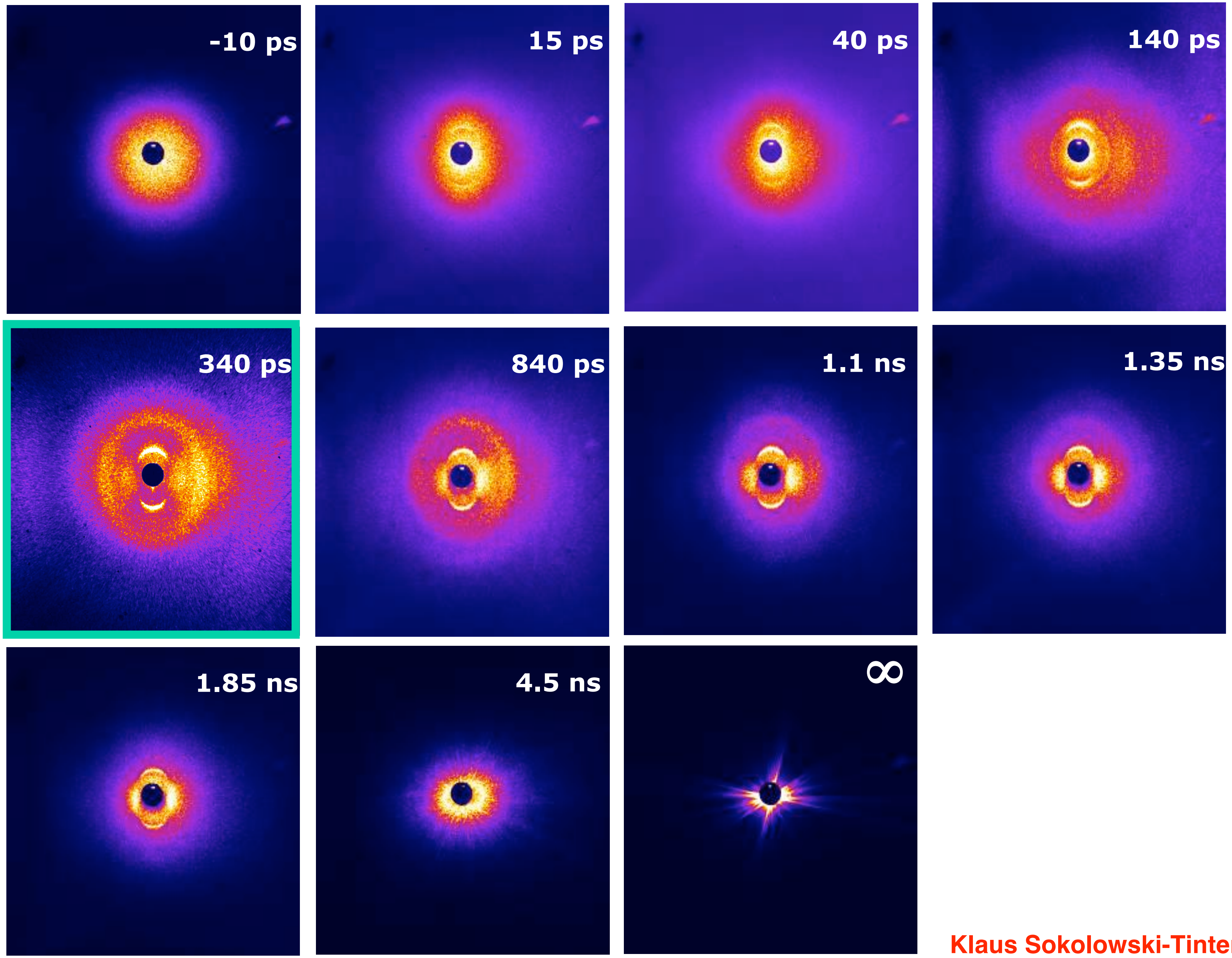
- Temperature and density at end of the FLASH pulse



Investigating structural changes on laser-irradiated surfaces using FLASH

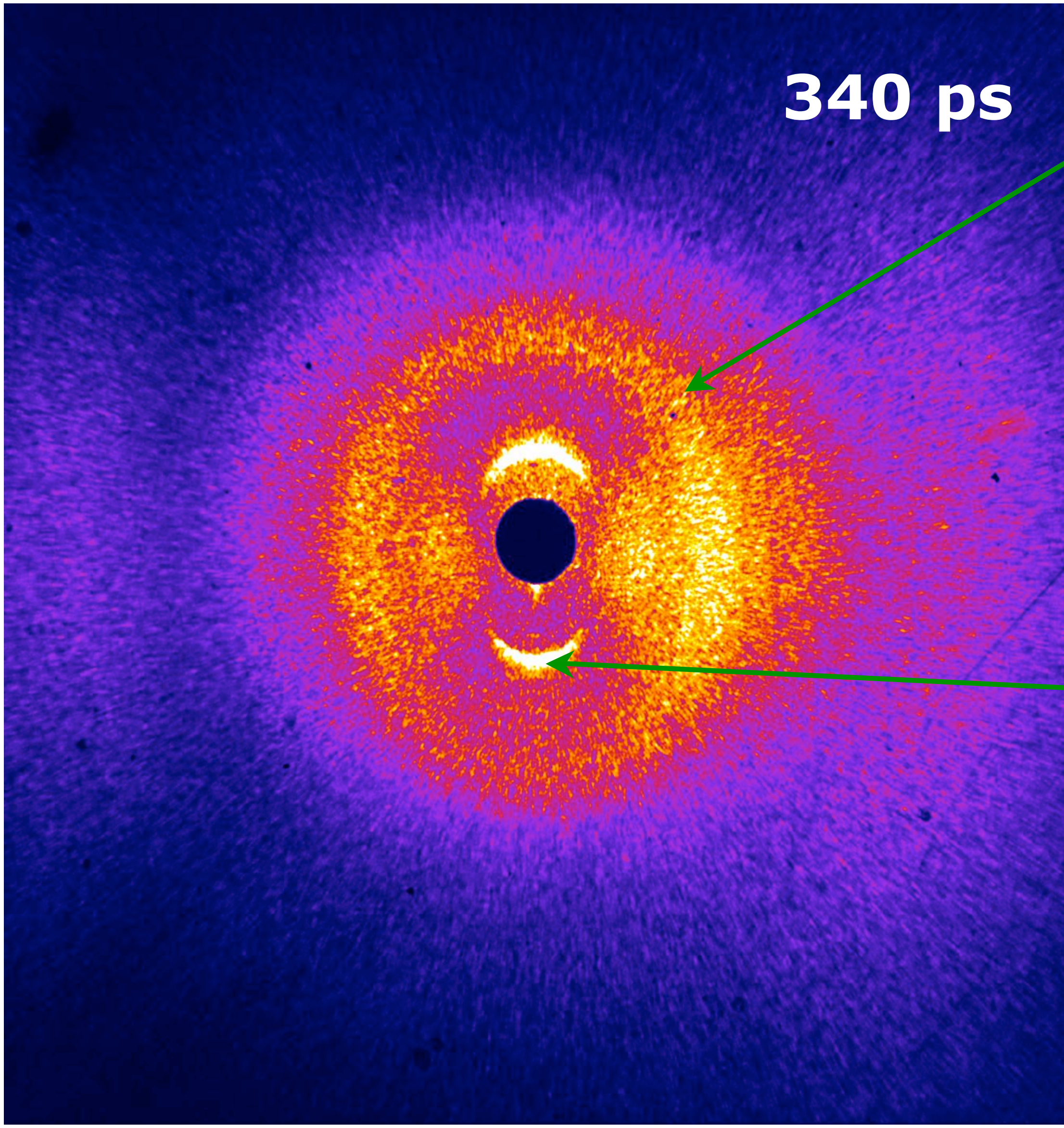


Klaus Sokolowski-Tinten (Essen)

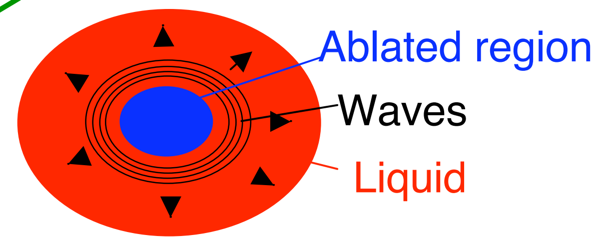


Klaus Sokolowski-Tinten

Scattering pattern provides information on formation of melting process



- **Surface waves**



- Initial calculations show ~150 nm waves

- **Ripples are LIPSS**
  - Measured 305 nm
  - Theory:

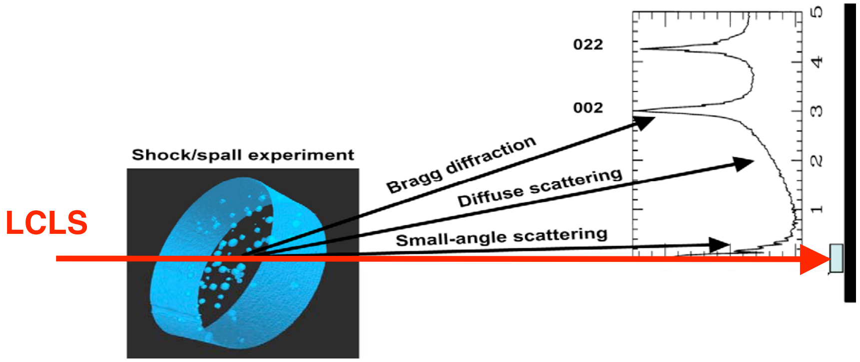
$$\Lambda = \lambda / (1 + \sin\theta)$$

$$\lambda = 527\ nm;\ \theta = 47^\circ$$

$$\Rightarrow \Lambda = 304\ nm$$

Klaus Sokolowski-Tinten

XFEL will probe WDM & high pressure states



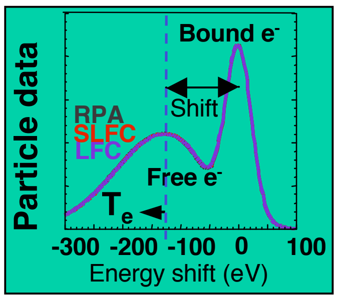
Standard techniques

- Bragg ⇒ *lattice compression and phase change*
- Diffuse ⇒ *dislocation content and lattice disorder*
- Small-angle ⇒ *sub-micron defect scattering*

- Short pulse freezes motion
- High flux yields single shot data
- High brightness probes “low-symmetry”
- High energy tunability accesses “thick” targets

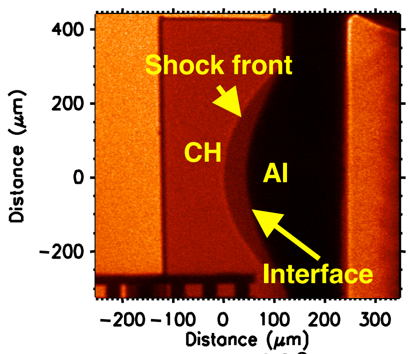
XFEL techniques

- X-ray Thomson Scattering ⇒ *T, N_e, γ* *(S. Glenzer, LLNL)*



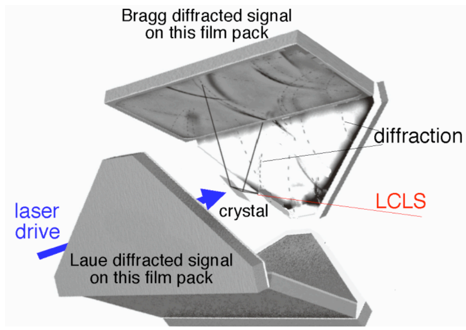
10^{13}

- Phase-Contrast imaging ⇒ *nm scale images* *(D. Hicks, LLNL)*



$< 10^{8}$

- Low Divergence ⇒ *nm-scale fs diffraction of real solids* *(J. Wark, Oxford)*



$\sim 10^{9}$

Warm Dense Matter and Free-Electron Lasers

A. Nelson[1], S. Toleikis[2], R. Sobierajski[3], P. Heimann[4], B. Nagler[5], M. Koslova[6], T. Whitcher[5], L. Juha[6], F. Khattak[7], J. Krzywinski[2], J. Wark[5], K. Sokolowski-Tinten[8], M. Fajardo[9], P. Zeitoun[10], P.Mercere[11], D. Riley[7], T. Tschentscher[2], R. Faeustlin[2], D. Schneider[4], T. Schenkel[4], S. Bajt[1], H. Chung[1], S. Moon[1], H. Scott[1], H. Chapman[1], …, R. W. Lee[1]

[1] Lawrence Livermore National Laboratory, Livermore, CA USA

[2] Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany

[3] Instytut Fizyki Polskiej Akademii Nauk, Warsaw, Poland

[4] Lawrence Berkeley National Laboratory, Berkeley, USA

[5] Oxford University, Oxford UK

[6] Academy of Sciences, Prague, Czech Republic

[7] Queen's University of Belfast, Belfast UK

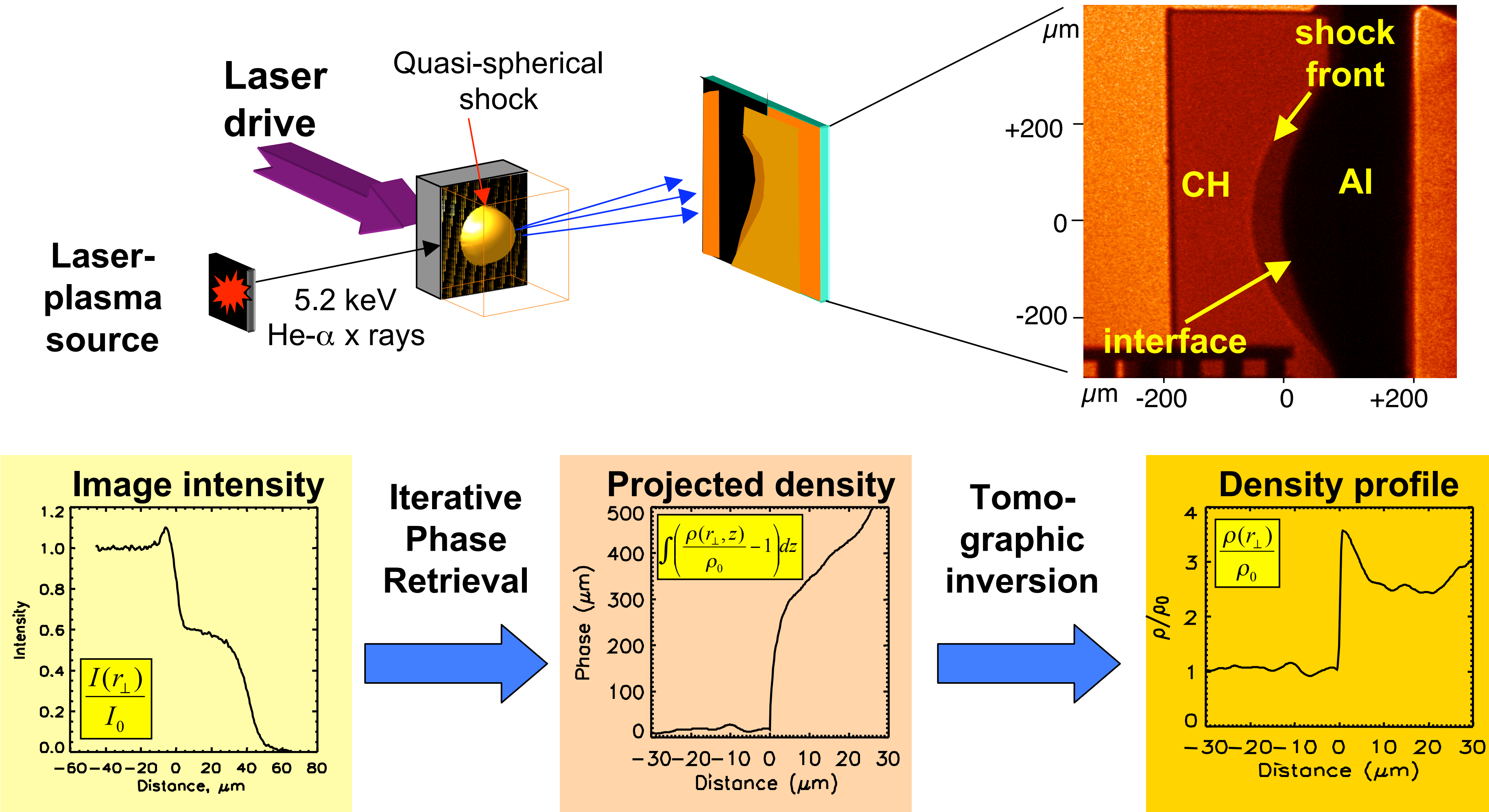
Universitat Duisburg Essen , Duisberg, Germany

[9]Instituto Superior Tecnico, Lisbon Portugal

[10]ENS Techniques Avancées, Paris, France

[11]Synchrotron Soleil, Gif-sur-Yvette, France

Current x-ray *phase-contrast imaging* at ~ 5 μm resolution uses laser-plasma sources

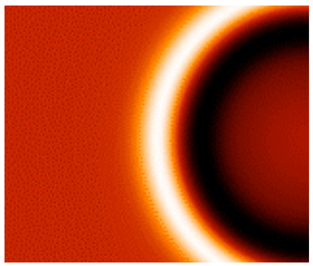


Current techniques are limited by spatial coherence & flux of laser-plasma x-ray source (D. G. Hicks 2006)

LCLS will enable coherent diffractive x-ray microscopy at the nanoscale

Shock front @ 2 μm resolution

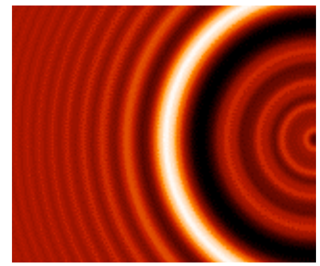
Phase-contrast imaging (near-field)



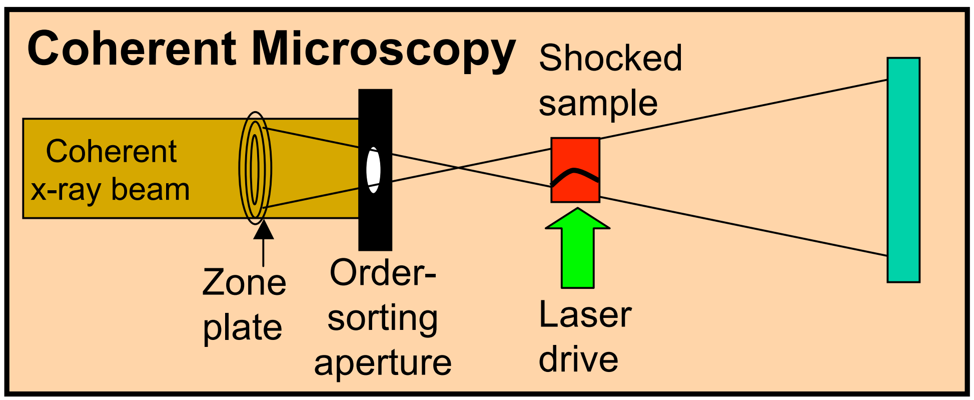
Coherent XFEL beam

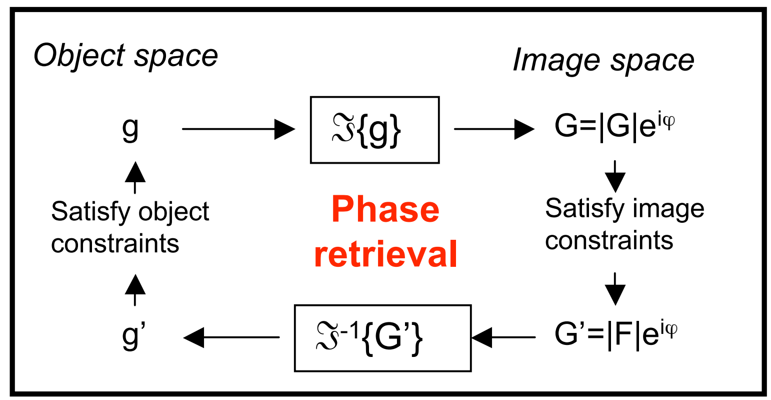
Single-pulse imaging: > 10^7 photons/pulse

Shock front @ 50 nm resolution



Coherent-diffractive imaging (far-field)



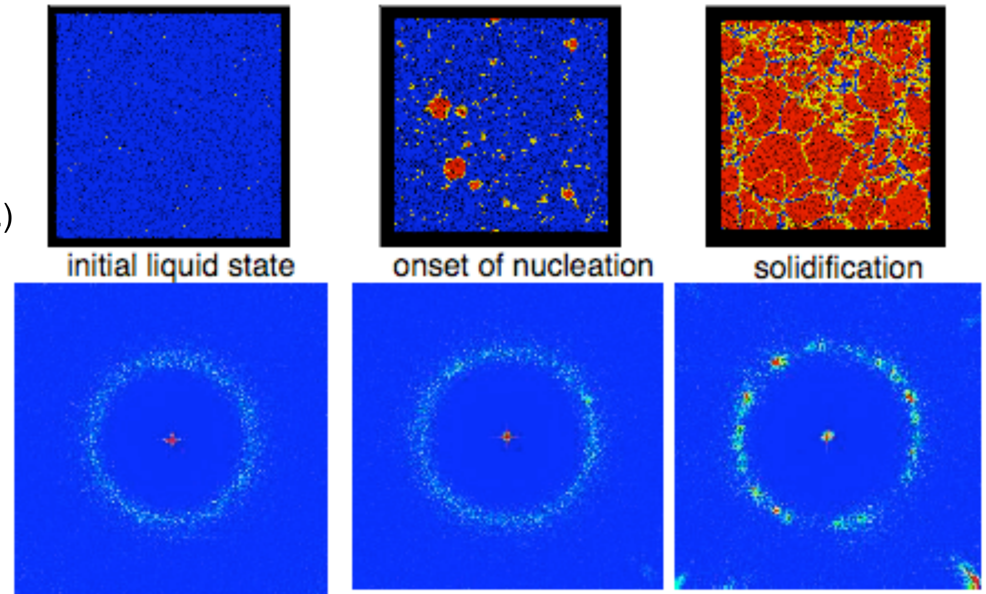


Dynamic processes on the nanoscale: shock front size (viscosity), phase transition kinetics, nucleation & growth, grain structure deformation

MD simulations of Ta show nucleation and solidification the XFEL can probe

MD simulation

(Streitz et al.)



initial liquid state | onset of nucleation | solidification

50 nm cube
16M atoms

LCLS diffraction patterns

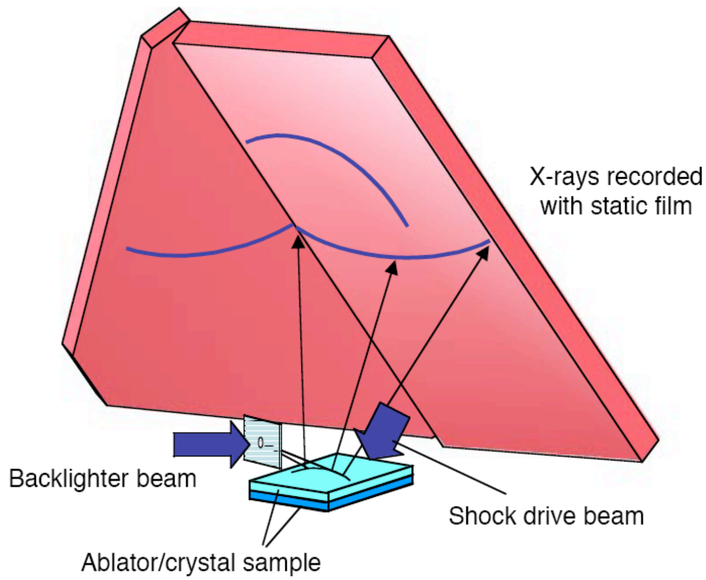
(Belak)

Uniform speckle | Non-uniformity of speckle | Higher intensity spots ⇒ number, size, & form of clusters

- Goal of *in situ* x-ray diffraction of shocked solids at granular level is to understand the microscopic to inform mesoscopic, and then macroscopic
- Study how individual grains respond elastically and plastically to high pressure as a function of orientation with respect to a given uniaxial shock wave
- XFEL is ideally suited to probe via diffraction polycrystalline high pressure solids because it is an ultra-bright, non-diverging, monochromatic source.

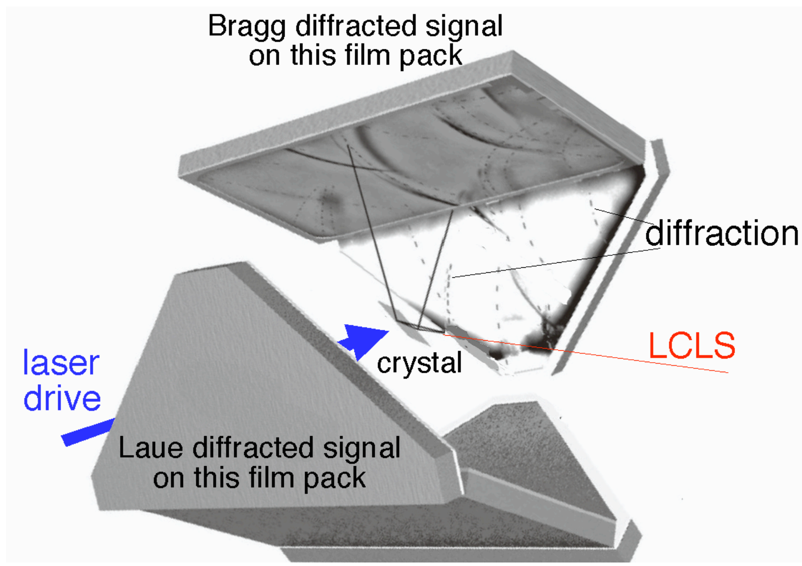
High-energy lasers provide shocks *and* high divergence x-ray probe

- **Schematic of Omega Laser shock experiment**



- Laser creates a shock in a mono-crystalline sample
- Delayed beams create ns-scale divergent source
- Angular spread of the x-ray sample from many crystal planes
- **Technique provides critical data on dynamics at high pressure**

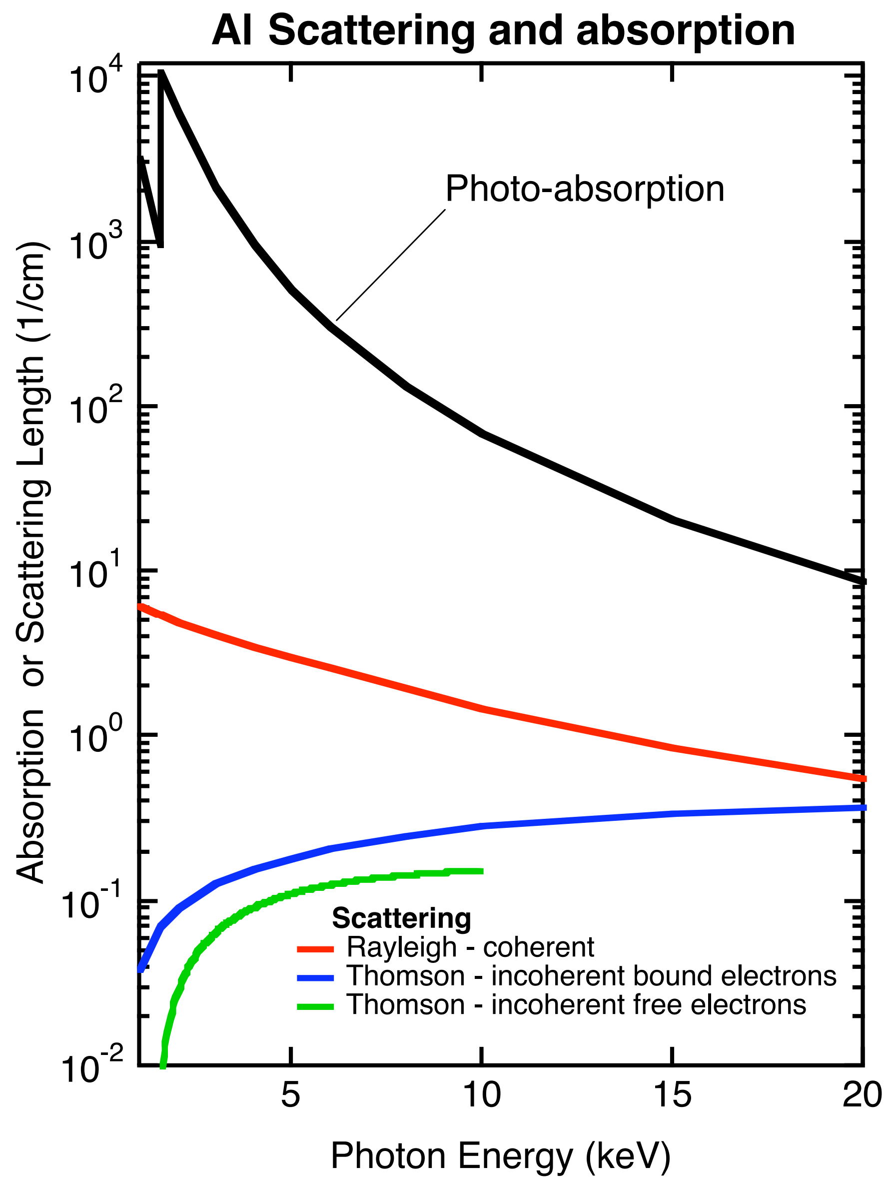
- **Schematic of LCLS XFEL shock experiment**



- Laser creates a shock in a polycrystalline sample
- XFEL create fs-scale non-divergent monochromatic source
- Grains in the polycrystal diffract the beam
- **Low Divergence ⇒ *nm-scale fs diffraction of real solids***

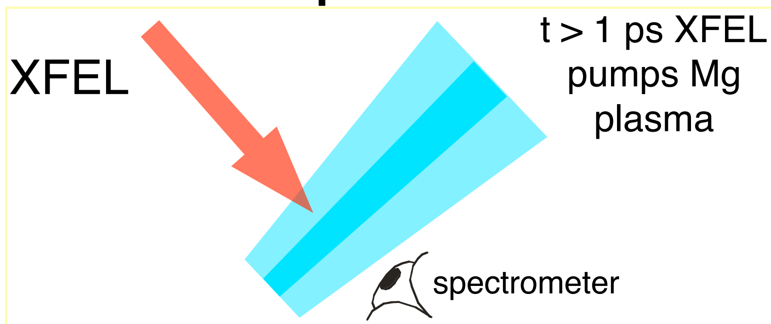
XFEL via photoionization pumping provides interesting WDM diagnostic potential

- Scattering from free electrons provides a measure of the T_e, n_e, $f(v)$, and plasma damping (S. Glenzer)
  - ⇒ structure alone ***not*** sufficient for plasma-like matter
- Due to absorption, refraction and reflection neither visible nor laboratory x-ray lasers can probe high density
  - ⇒ little to no high density data
- FEL signals will be well above noise for all HED matter



XFEL provides an opportunity for HEDS plasma spectroscopy

- Source for hollow ion experiment



- Study the $K^0L^yM^z \rightarrow K^1L^{y-1}M^z + h\nu_{emitted}$

- **Photoionization of multiple ion species**:

  $K^xL^yM^z + h\nu_{XFEL} \rightarrow K^{x-1}L^yM^z + e$ (x=1,2; y=1-8; z=1,2)

- **Auger Decay of multiple ion species:**

  $K^xL^yM^z + h\nu_{XFEL} \rightarrow K^{x-1}L^yM^z + e \rightarrow K^xL^{y-2}M^z + e$

- **Sequential multiphoton ionization:**

  $K^xL^yM^z + h\nu_{XFEL} \rightarrow K^{x-1}L^yM^z + e + h\nu_{XFEL} \rightarrow K^0L^yM^z + e + h\nu_{XFEL}$

  $\rightarrow K^0L^{y-1}M^z + e + h\nu_{XFEL} \rightarrow \ldots$

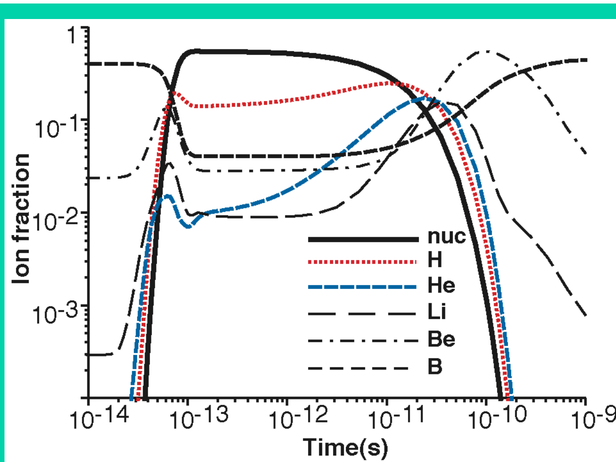
  $K^xL^yM^z + h\nu_{XFEL} \rightarrow K^{x-1}L^yM^z + e + h\nu_{XFEL} \rightarrow K^{x-1}L^{y-2}M^z + 2e$

- **Direct multiphoton ionization:**

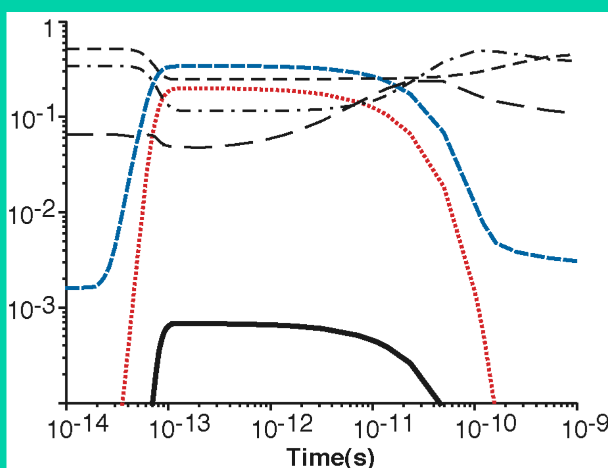
  $K^xL^yM^z + 2h\nu_{XFEL} \rightarrow K^0L^yM^z + 2e$

Hollow ion studies in the HED regime will yield data on kinetic processes and diagnostics

- LCLS will create unique states of matter and provide *first* hollow ions
  - Simulations: 5×10^{10} photons in 100 fs, 30 μm spot into a $n_e = 10^{21}$ cm^{-3} Mg plasma

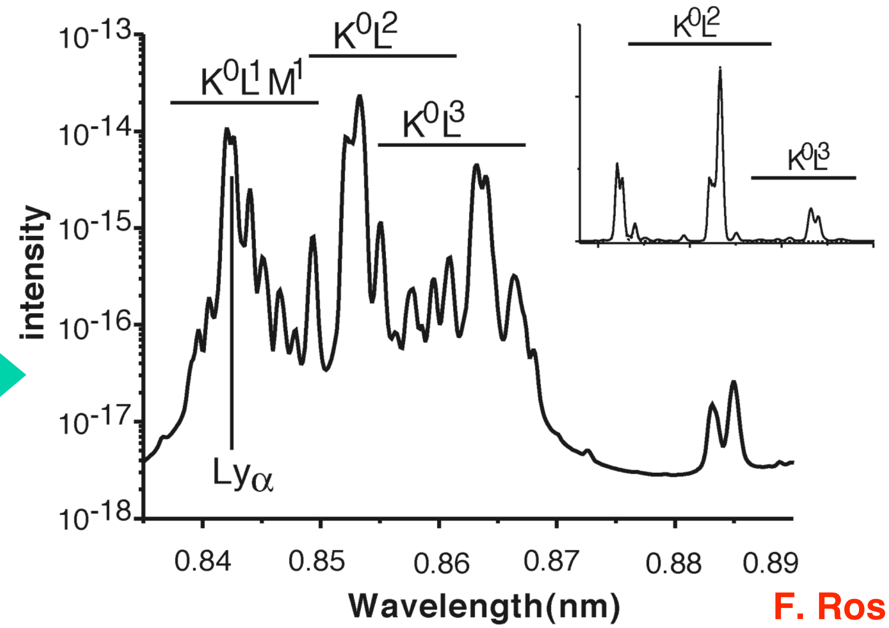


- 3.10 keV ionize to bare nucleus in < 50 fs



- At 1.85 keV maximize He-like state

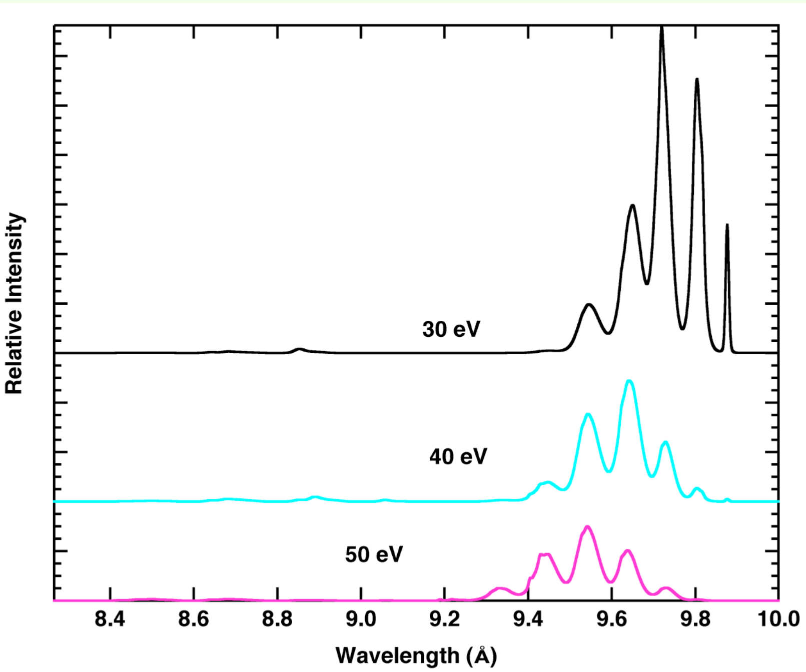
- Recombination kinetics take > 10 ps
- Time-integrated spectrum shows dominance of hollow ion emission



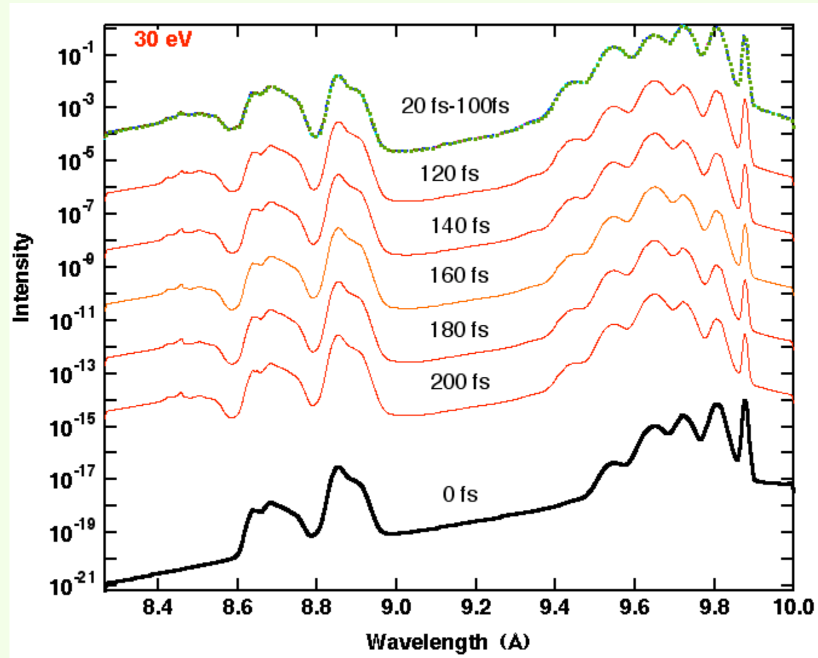
F. Rosmej

In Warm Dense Matter regime the hollow ions provide time-resolved diagnostic information

- XFEL forms unique states ***and*** provides *in situ* diagnostics with ~100 fs resolution
  - 5×10^{10} 1.85 keV photons in 30 μm spot into a **n_e=10^{23} cm^{-2}** plasma
  - Strong coupling parameter, Γ_{ii} = Potential/Kinetic Energy ~ 10



• Spectra vary with temperature



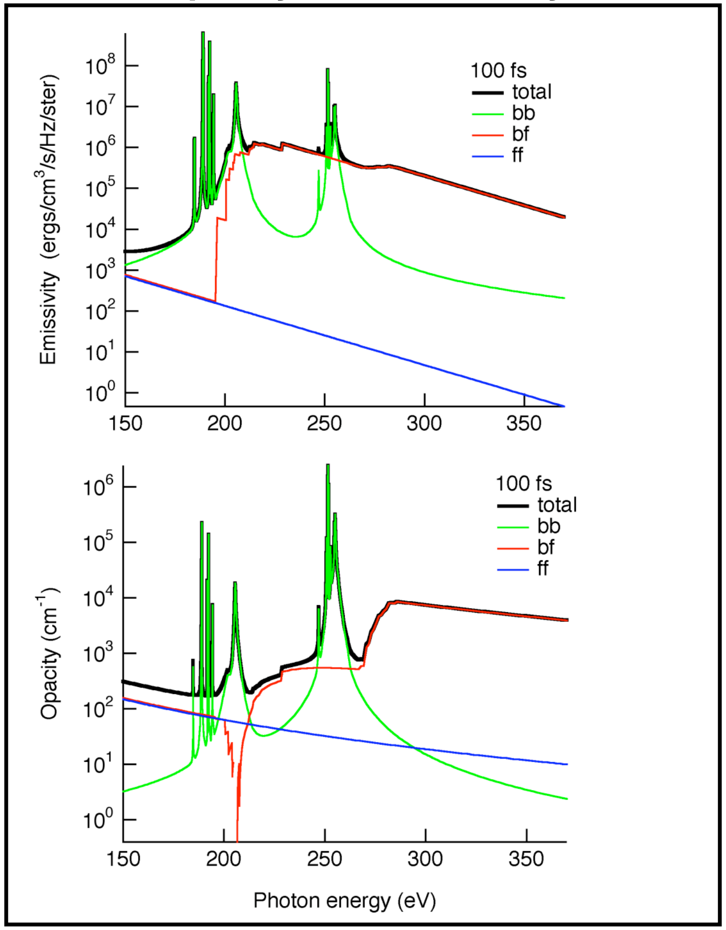
• At high n_e emission lasts ~100 fs

H.-K. Chung

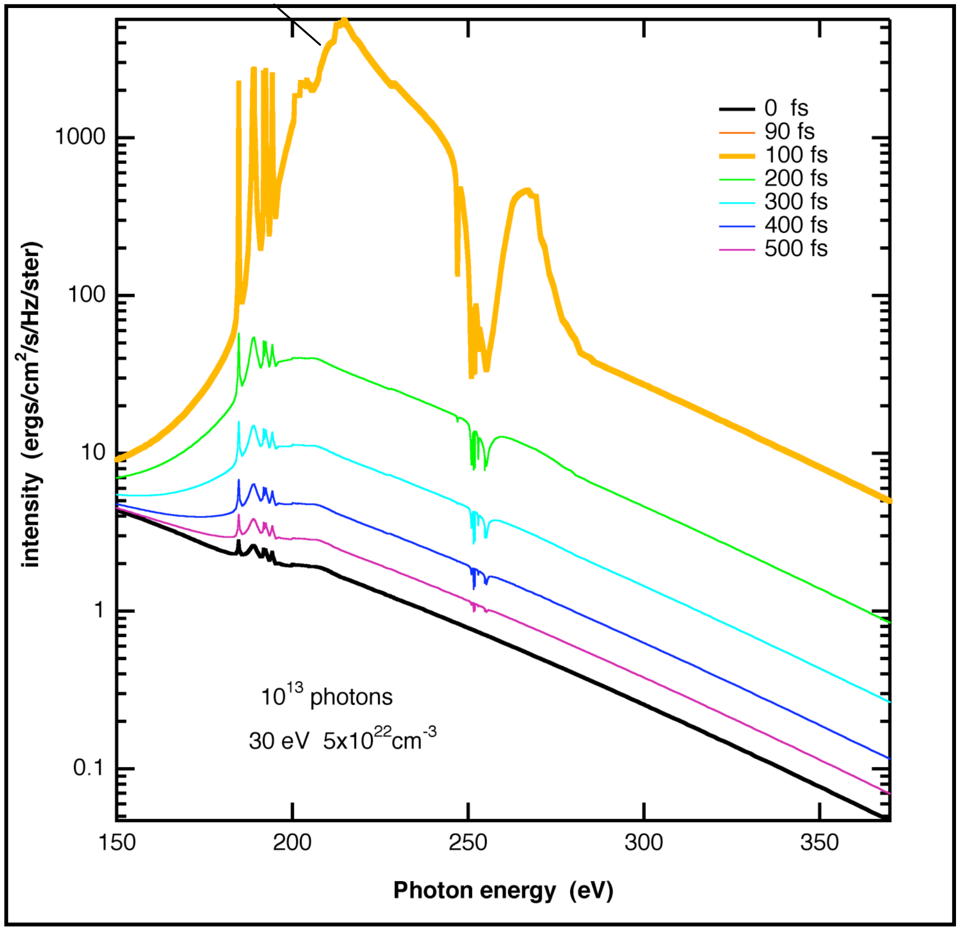
Finally, saturating the continuum using FLASH may provide a ~100 fs absorption source

- He-like B plasma at 30 eV, $5x10^{22}$ cm^{-3}, 1 mm in length
- FEL tuned to H-like boron 1-2 transition at 250 eV

Opacity and Emissivity



Continuum rises rapidly and last for ~100 fs



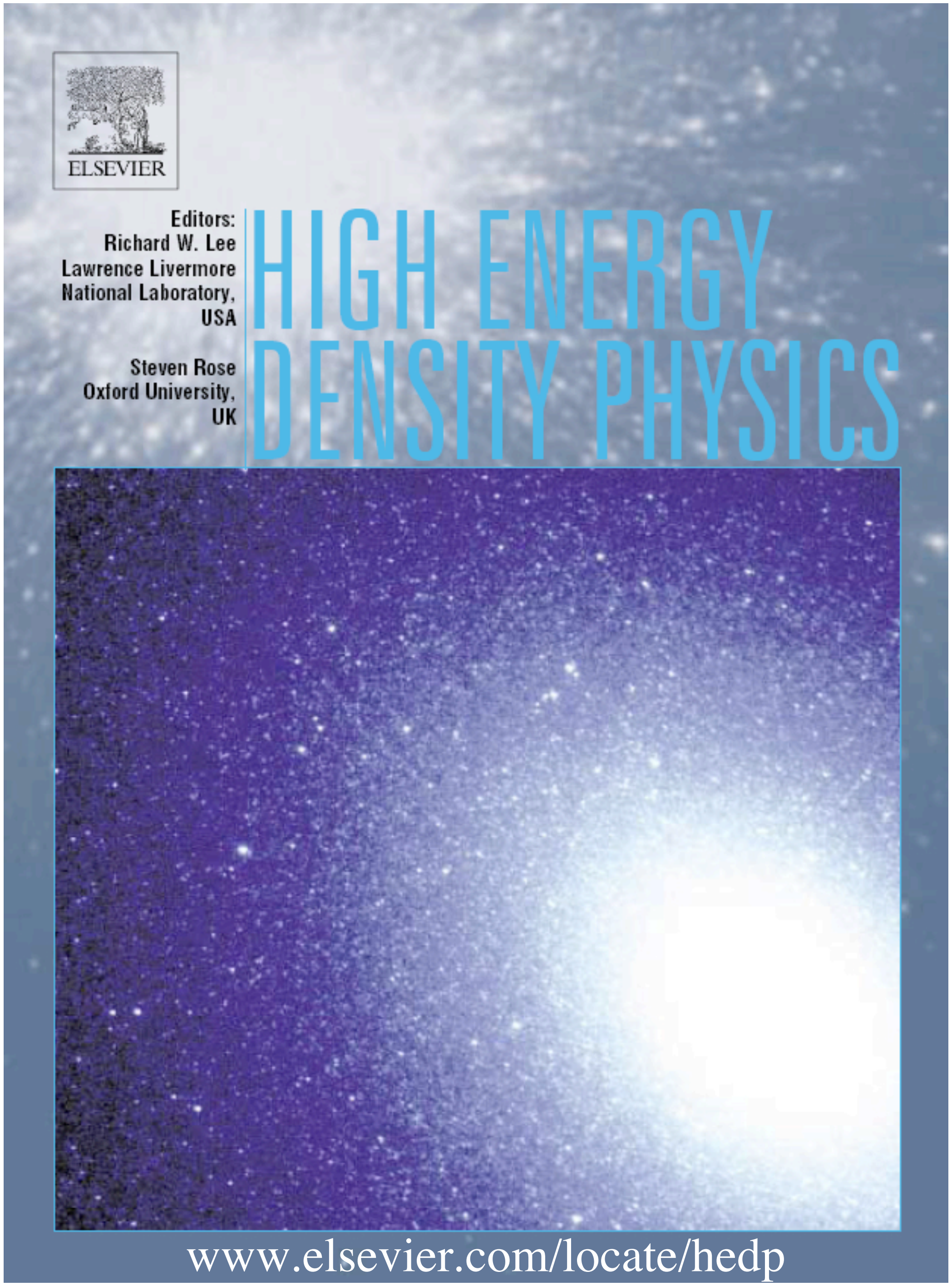
H.-K. Chung

Summary of HEDS using sub-ps intense x-ray sources - XFELs

- For both the hot and warm dense matter regimes new possibilities opened up by XFELs

- For WDM the FELs provide
  - WDM creation: Fast uniform heating source
  - WDM diagnostics: Thomson Scattering, K_{α} temperature measurement, sub-ps absorption sources, phase contrast imaging, diffraction for high pressure states

- For HDM the FELs provide:(not shown)
  - Fast deposition may create hot, high pressure matter
  - Plasma spectroscopic probes of kinetic and radiative processes
  - Diagnostic potential: Thomson scattering



High Energy Density Physics is an international journal covering original experimental and related theoretical work studying the physics of matter and radiation under extreme conditions. 'High energy density' is understood to be an energy density exceeding ~10^{11} J/m^3. The editors and the publisher are committed to provide this fast-growing community with a dedicated high-quality channel for their original findings.

Papers suitable for publication in this journal cover topics in both the ***warm and hot dense matter regimes***, such as laboratory studies relevant to non-LTE kinetics at extreme conditions, planetary interiors, astrophysical phenomena, inertial fusion and includes studies of, for example, material properties and both stable and unstable hydrodynamics. Developments in associated theoretical areas, for example the modelling of strongly coupled, partially degenerate and relativistic plasmas, are also covered.

Motivations for *High Energy Density Physics*

- **Cross-disciplinary studies can be aired**
  - In HEDP, a research area incorporating several fields, it is essential to provide a forum for discussion.

- **Venue for the discussion of innovative ideas**
  - With the rapid expansion of HEDP research into new experimental and theoretical areas is critical.

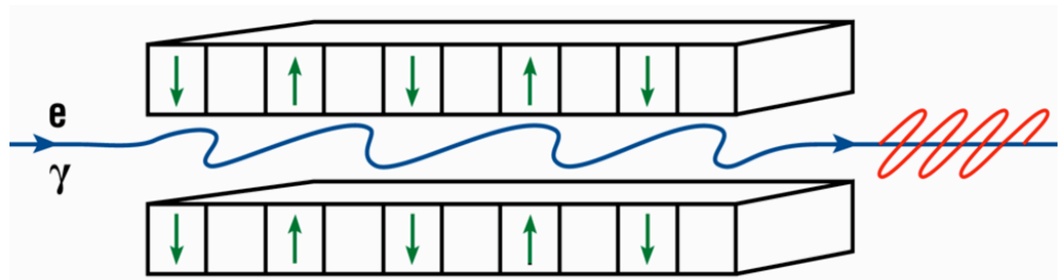
- **Promotion of the broad-based interests of the field**
  - With its boundaries still being defined, *HEDP* can assist in focusing attention on issues related to "definition".

  - Papers can delve into why, *e.g.*, certain strongly coupled systems are not interesting, while others with similar strong-coupling parameters are intriguingly difficult.

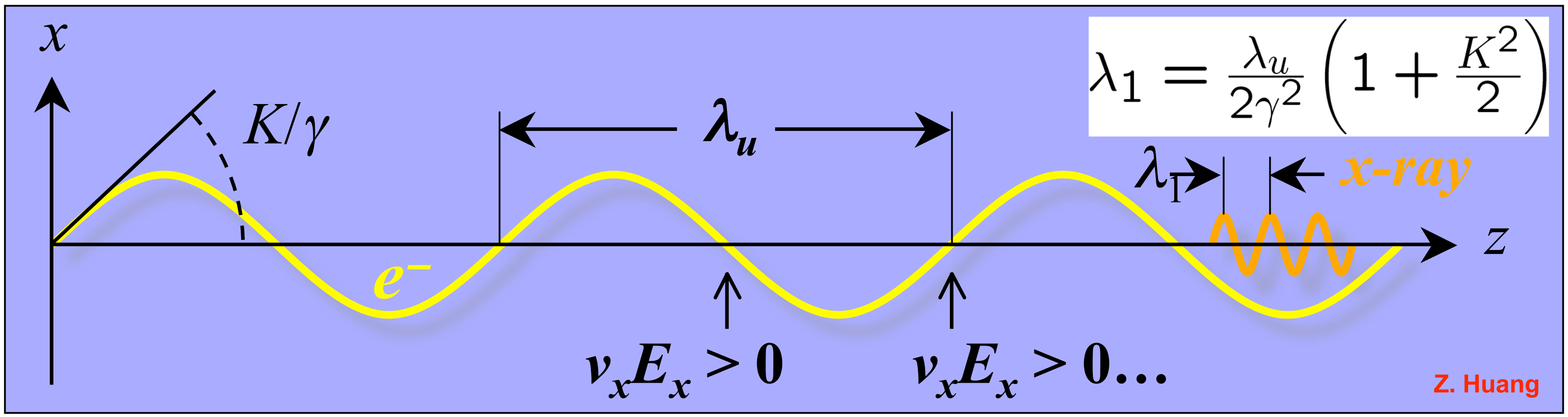
- **Provision of a scientific outlet for rejuvenation**
  - Older methods, when applied to new HEDP processes, can become important again and *HEDP* is an scientific outlet for that rejuvenation

The End

FEL Principles



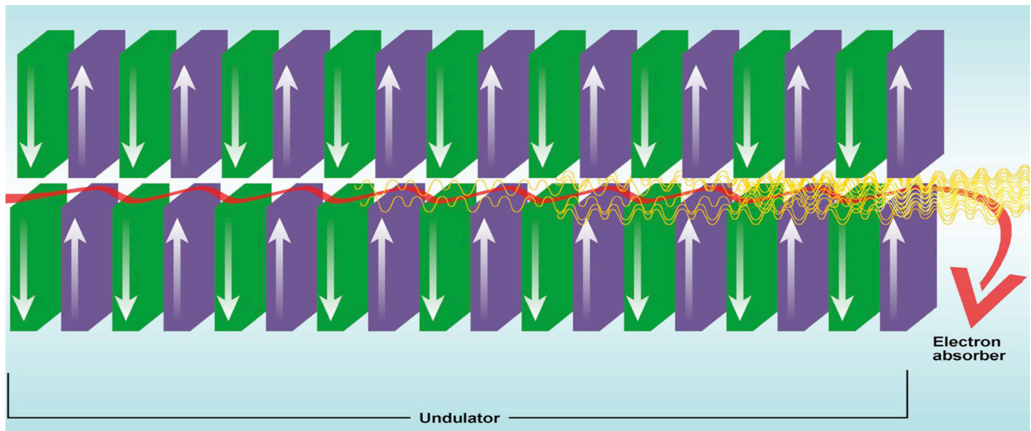
- Electrons slip behind EM wave by λ_1 per undulator period (λ_u)



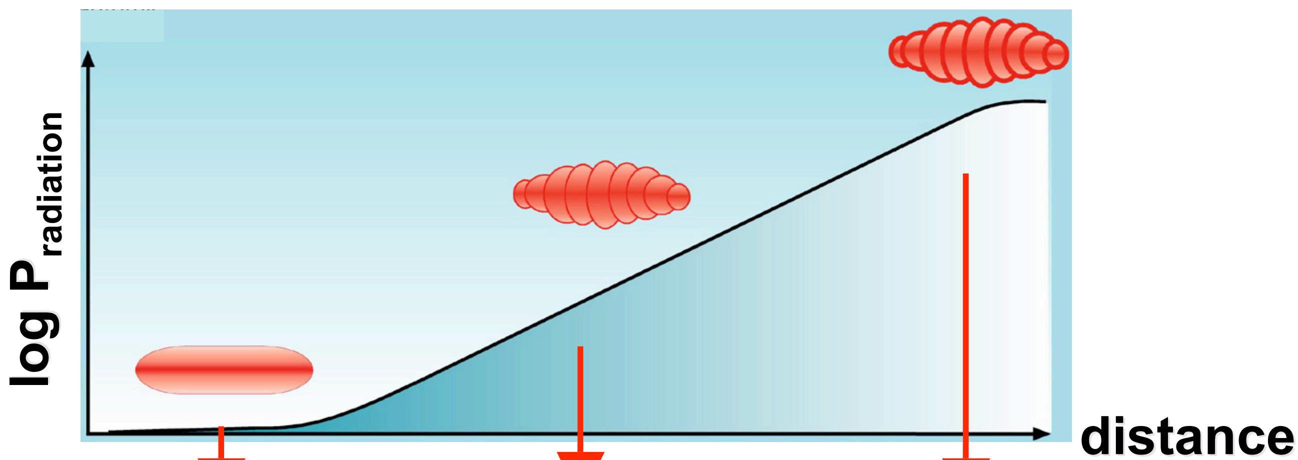
- Due to sustained interaction, some electrons lose energy, while others gain ➔ energy modulation at λ_1 
- e^- losing energy slow down, and e^- gaining energy catch up ➔ density modulation at λ_1 (microbunching) 
- Microbunched beam radiates coherently at λ_1, enhancing the process ➔ exponential growth of radiation power

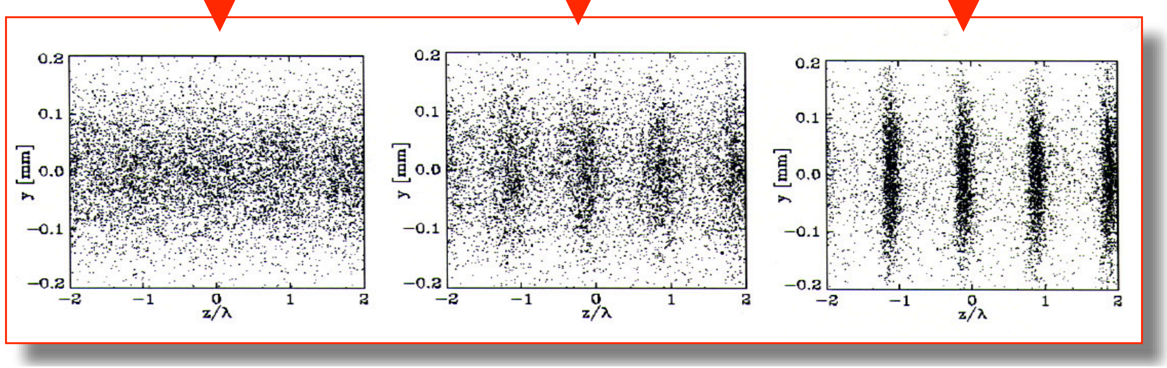
Self-Amplified Spontaneous Emission (SASE)

e^- beam



photon beam

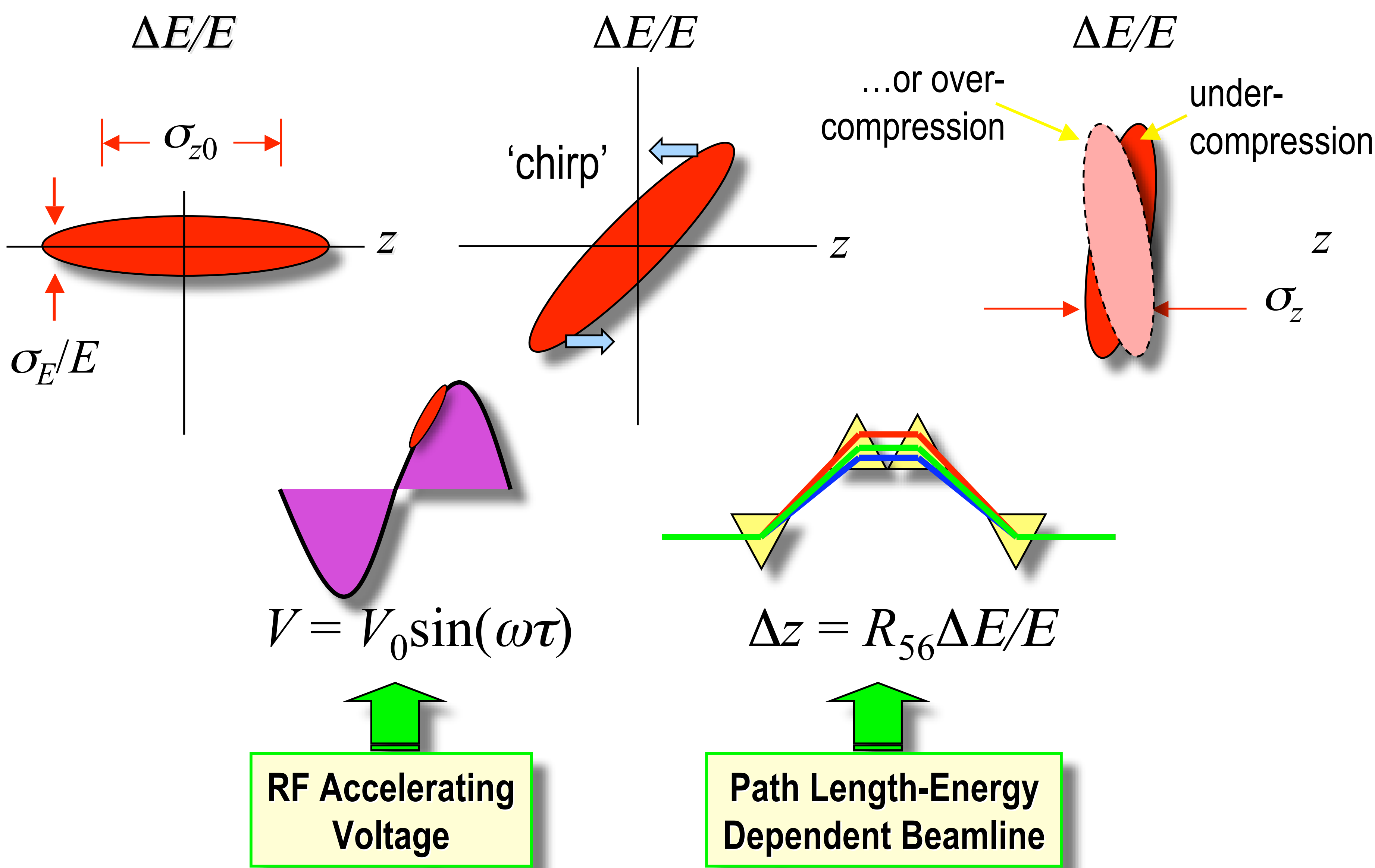




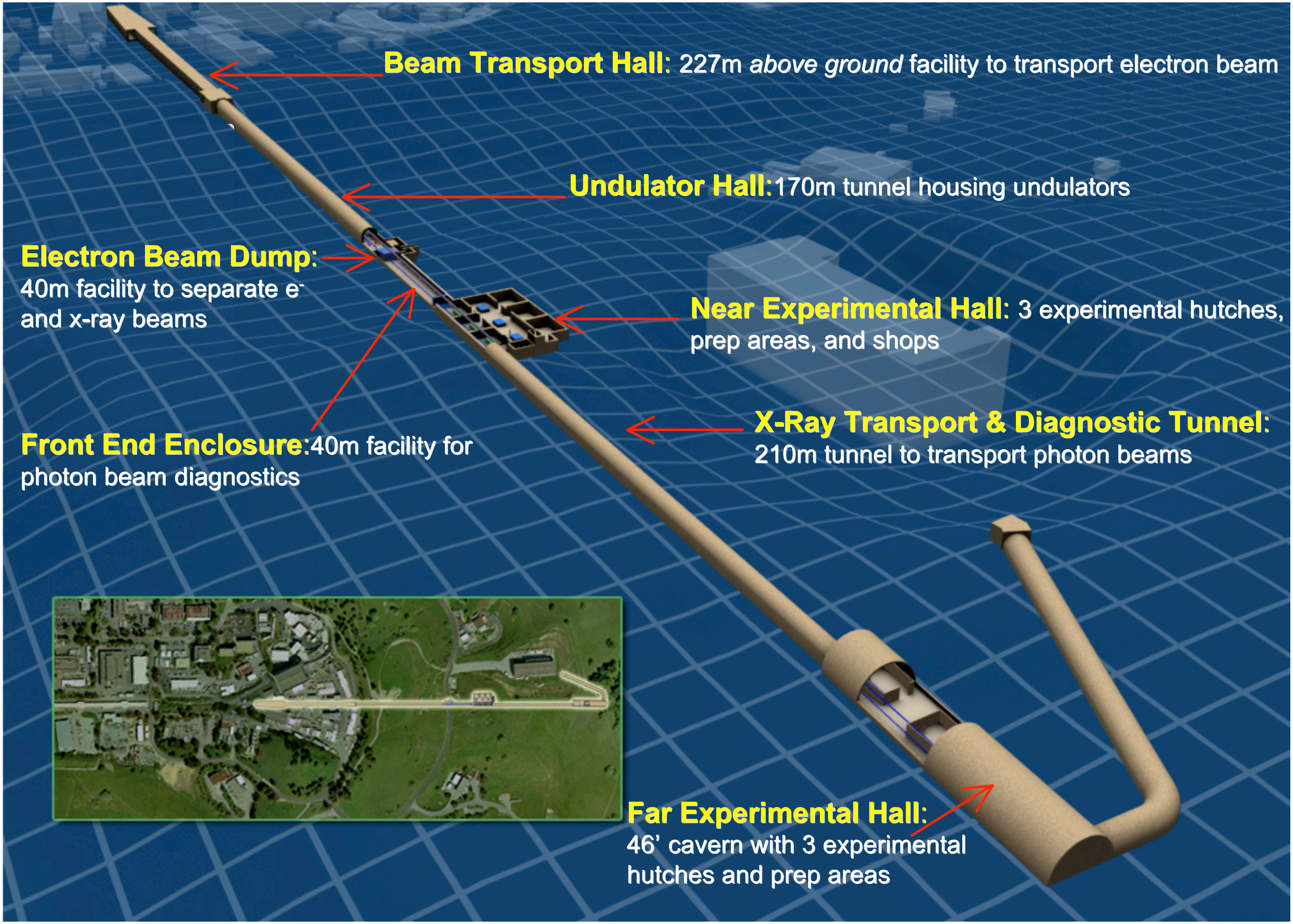
From DESY

e^- beam is microbunched at the radiation wavelength

Magnetic Bunch Compression

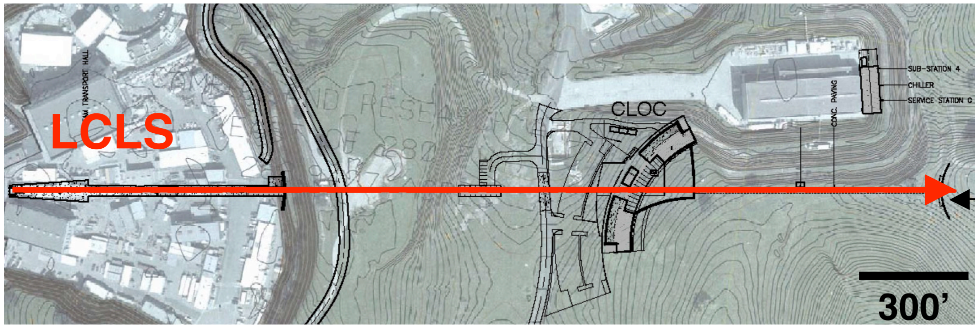


Beam Transport from Linac Through X-Ray Halls

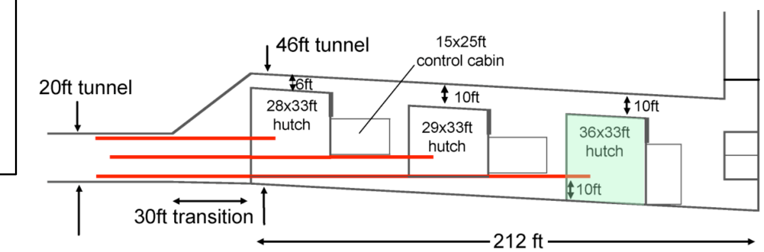


LCLS HEDS end station will house high energy and short pulse lasers

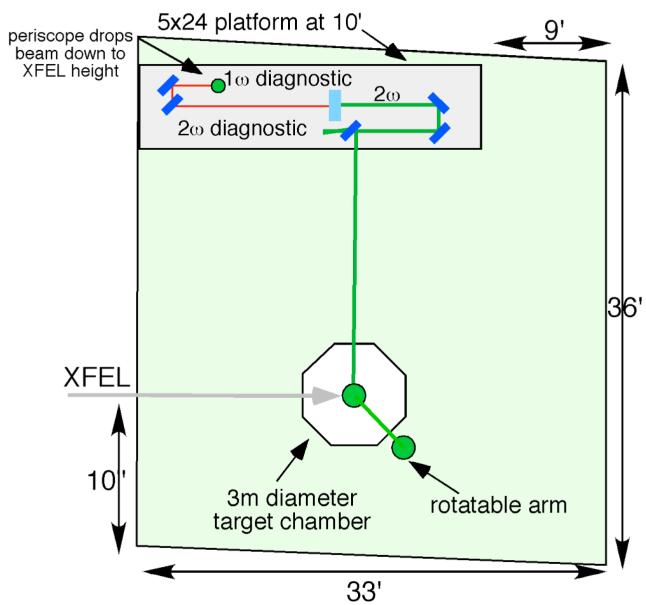
Aerial view of SLAC



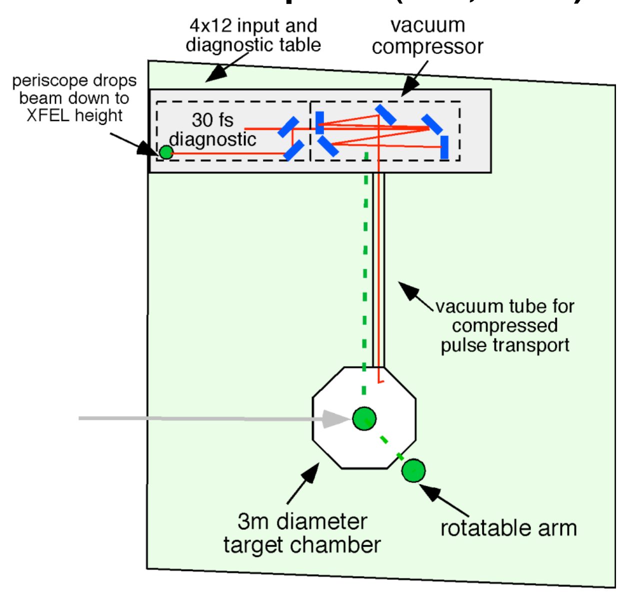
Far Experimental Hall



Long-pulse (~100J, 5ns)



Short-pulse (≥1J, 30 fs)



- Lasers can be transported from mezzanine above into hutch